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**National University of Ireland, Cork**



**Tribology of malt-based beverages: Development and  
application of method**

Thesis presented by

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**School of Food and Nutritional Sciences**

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## **Declaration**

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Daniel Fox

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*"In the time of your life, live – so that in that wondrous time you shall not add to the sorrow and the misery of the world, but shall smile at the infinite delight and mystery of it."*

*William Saroyan, 1939*

## Abstract

Soft tribology, i.e. the measurement of friction as a function of speed between two compliant surfaces, has found applications in food science and there is a growing body of theoretical and practical knowledge of fundamental mechanisms of lubrication as well as increasingly strong correlations between tribology and sensory data. Soft tribology is generally conducted using either commercially or in-house built tribometers however, the recent decade has seen a rise in the use of rheometers with tribology attachments. Based on current literature, knowledge gaps and potential avenues for future research have been identified. These include investigations on hydrophobicity of surfaces, surface wear (running-in), cleaning procedure of the attachment and tribopairs, speed (range and method of increase/decrease) and measuring system configuration.

In the current research, frictional parameters of 10 beers (5 alcoholic and their non-alcoholic counterpart) were measured using an Anton Paar MCR301 rheometer with a tribology attachment (BC12.7, Anton Paar, Graz, Austria), and a range of variables was extracted and subjected to dimension reduction analysis (Principal Component Analysis, clustering, and correlation analysis). Sensory data consisting of a numeric mouthfeel rating and written reviews from an online beer-rating website ([www.ratebeer.com](http://www.ratebeer.com)) were compiled, transformed, and correlated with the tribology data. Based on Frictional parameters of the beers, clear differences were observed between alcoholic and non-alcoholic beers, as well as those beers with high or low mouthfeel rating. Text-mining and clustering of the written reviews led to the development of 7 overall sensory descriptors; "watery", "smooth", "thick", "bitter", "foam", "astringent", and "sour", related to mouthfeel. Frictional parameters related to the static (speed range  $\sim 10^{-8}$ - $10^{-5}$  m/s), boundary (speed range  $\sim 10^{-5}$ - $5 \times 10^{-5}$  m/s) and beginning of the mixed regime (speed range  $\sim 5 \times 10^{-5}$ - $10^{-4}$  m/s) were correlated with "watery", "smooth", and "thick", while "bitter", "foam", "astringent", and "sour" were represented later in the mixed regime (speed range  $\sim 10^{-4}$ - $10^{-3}$  m/s).



$4 \cdot 10^{-3}$  m/s). These results are significant in two ways; firstly, they indicate the usefulness of online beer reviews as a means to gather reliable sensory data, and secondly, they demonstrate tribology as a tool to instrumentally define and determine important mouthfeel parameters of beer. Further research is needed to fully validate this methodology; results from the online database should be compared to the outcome of a consumer panel under controlled settings, and a wider range of beers of different styles should be tested to fully understand the correlations between sensory phenomena and frictional parameters.

# Chapter I Tribology using rheometers: Literature review

## 1 Introduction

“Mouthfeel” is a self-explanatory term used to describe a complicated and multi-faceted sensory concept: it encompasses the tactile sensations experienced during mastication. However, going beyond “good” and “bad” and separating those sensations into meaningful terms and correlating them with instrumental measurements requires a developed palate, vocabulary and methodology. The mouthfeel of a given product is an important determinant of the liking and acceptance by consumers (Guinard & Mazzucchelli, 1996), and product developers of food and beverages, referred to collectively as food in this review, are therefore often interested in measuring mouthfeel in order to optimise this important parameter comprising a wealth of different sensory phenomena. Mouthfeel can be defined as the “... tactile (feel) properties perceived from the time at which solid, semi-solid or liquid foods or beverages are placed in the mouth until they are swallowed” (Guinard & Mazzucchelli, 1996). Currently, trained sensory panels are used to determine mouthfeel; however, these are expensive and time-consuming, especially when dealing with large sample-sizes (Prakash, 2016). For certain foods, i.e. semi-solid to liquid, the flow-characteristics, rheological, have been successfully correlated with certain mouthfeel attributes; e.g. stickiness, thickness and mouthcoating (He, Hort, & Wolf, 2016), and, while rheological measurements in some cases can distinguish between samples with differences in mouthfeel, viscosity alone often fails to accurately explain the complex phenomenon of oral physical interactions that constitutes mouthfeel (Prakash, Tan, & Chen, 2013; Selway & Stokes, 2014). This shortcoming can be explained by the fact that rheology is a property principally related to the substrate, i.e. the food, while mouthfeel arises from physical and chemical interactions between the food and oral cavity that cannot be described solely by the flow-characteristics of the food in question (Selway & Stokes, 2014). This is

not to say that bulk properties such as viscosity and density are not important parameters influencing mouthfeel. As such, development of a high-throughput, inexpensive and reproducible method would offer advantages to both industry and academic researchers. In the last decades considerable effort has been put into development of such a system by food scientists. A promising method is the use of tribology which can be succinctly defined as follows: “The science of tribology principally involves studying the characteristics of the film situated between contacting bodies and the consequence of its failure or absence” (Stokes, 2012). The word tribology is a contraction of the Greek root words for “rubbing” (tribo) and “study of” (logia). “Soft” tribology can further be defined as the study of lubrication and friction using compliant (i.e. deformable) surfaces to better mimic the conditions in the oral cavity (Joyner, Pernell, & Daubert, 2014a; Rudge, Scholten, & Dijkman, 2019).

The term tribology was coined in 1966 (Jost, 1966) but the study of friction reaches far back in history. Development of more sensitive measuring systems and advances in polymer-science to produce surfaces that mimic biological systems; i.e. deformable surfaces with defined wetting characteristics, has led to the emergence of bio-tribology or “soft” tribology, a branch of tribology that studies the frictional properties of biological systems. Examples of applications include prosthetics (Samaroo et al., 2017; Stevenson et al., 2019; Voutat, Nohava, Wandel, & Zysset, 2019) , contact lenses (Pitenis et al., 2017), cosmetics (Timm, Myant, Spikes, & Grunze, 2011), dentistry (Cai, Li, & Chen, 2017), medicine (Batchelor, Venables, Marriott, & Mills, 2015) and more, as well as the study of friction during oral processing (Sarkar, Andablo-Reyes, Bryant, Dowson, & Neville, 2019).

The last decade has seen an increased use of tribo-attachments to rheometers rather than tribometers such as the Mini Traction Machine (MTM, PCS instruments, UK), commonly used for tribological measurements (Shewan, Pradal, & Stokes, 2019). Rheometers are generally ubiquitous in academic food science labs and often present in large food companies (although they may be out of the price-range for

small to medium sized companies) and offer precise normal force control as well as an increased speed range compared to many conventional tribometers; however, as noted recently by (Shewan et al., 2019), several disadvantages exist, namely: a lack of fundamental studies (as compared to the wealth of fundamental studies on tribometers), limitations in the movement profile; i.e. only rotational, limited knowledge on challenges related to interpretation and understanding of the output, and lack of reporting or consideration of surface wear (Sarkar & Krop, 2019; Shewan et al., 2019).

This review aims to introduce tribology to food scientists, assuming no prior knowledge of the area but a basic understanding of rheology and food physics/chemistry. The focus will be on comparison as far as possible of methodologies across categories of food and tribological attachments, with an emphasis on preparation protocols and measuring system parameter settings. Most tribology studies so far has focused on dairy-related products or hydrocolloid solutions (Sarkar & Krop, 2019; Shewan et al., 2019) however, measurements on beverages such as wine, tea and soft drinks are possible (see e.g. Chong et al., 2019; Laguna & Sarkar, 2017; Steinbach, Guthrie, Smith, Lindgren, & Debon, 2014) and as such, the field of tribology could well be extended to include other beverages (e.g. beer and other malt-based beverages). Fermented wort with its low viscosity; low concentrations of polysaccharides, proteins, polyphenols, and their complexes; presence of fermentation by-products; and hop-extracts presents a new challenge for tribologists, with many avenues worthy of exploration. For example, hops (*Humulus lupulus*) polyphenols and bitter acids play an important role in the perceived fullness, bitterness, astringency, and stickiness of beers (Goiris et al., 2014; Oladokun et al., 2016); these are sensory phenomena that have already been correlated with tribological measurements in other food systems (Sarkar & Krop, 2019) and so the relationship between these hop compounds and mouthfeel of beer could potentially be further elucidated using tribology. Another potential area of investigation is the effect of adjuncts on mouthfeel of beer; for example, recently the effect of arabinoxylans from unmalted rye on mouthfeel was investigated and was found to positively influence the perceived fullness

of beers (Langenaeken, De Schutter, & Courtin, 2020). Additionally, ways to instrumentally measure the mouthfeel of beers could help in the improvement of beers that are generally perceived as having poor mouthfeel, e.g. non-alcoholic beers (Bellut & Arendt, 2019; Krebs, Müller, Becker, & Gastl, 2019)

For reviews and articles dealing with the difference between rheology and tribology (Chen & Stokes, 2012), in depth introductions to the theoretical background of tribology (Sarkar, Andablo-Reyes, et al., 2019; Stokes, 2012), theoretical work on lubrication of soft viscoelastic surfaces (Pandey, Karpitschka, Venner, & Snoeijer, 2016) as well as linking tribology and sensory data (Sarkar & Krop, 2019; Shewan et al., 2019), and more complex modelling in tribology (Chen & Opara, 2013; Smith, Guthrie, Steinbach, Lindgren, & Debon, 2015; Vakis et al., 2018) the references cited here are recommended.

## 2 Fundamentals of soft tribology

### 2.1 Tribology is a system property

An important theoretical framework to be considered before embarking on tribological work is that tribology is a system property (Hutchings, Gee, & Santner, 2006). This means that careful consideration, especially when interpreting and comparing tribological data, should be given to the nature of

- a) The measuring system – e.g. type of machine, tribopair configuration and type of movement
- b) The surfaces – e.g. roughness, hydrophobicity, viscoelasticity
- c) The lubricant (food) – e.g. rheological properties, heterogeneity (emulsion, particle size, presence of gas, surface-active ingredients) (Sarkar & Krop, 2019; Shewan et al., 2019)

A wide array of parameters (figure 1) influence mouthfeel and play a role doing tribological measurements. An important implication of this is that, when performing a tribological measurement, the output (data) is not only a reflection (product) of the food-systems lubricating properties as affected by structural and compositional characteristics. It is also a measurement of the mechanical and surface properties of the surfaces used as affected by composition, production method, treatment before use,

humidity, temperature and other environmental factors as well as a result of the choice of measuring system, protocol and data-gathering strategy (Joyner et al., 2014a; Sarkar, Andablo-Reyes, et al., 2019; Sarkar & Krop, 2019; Shewan et al., 2019).

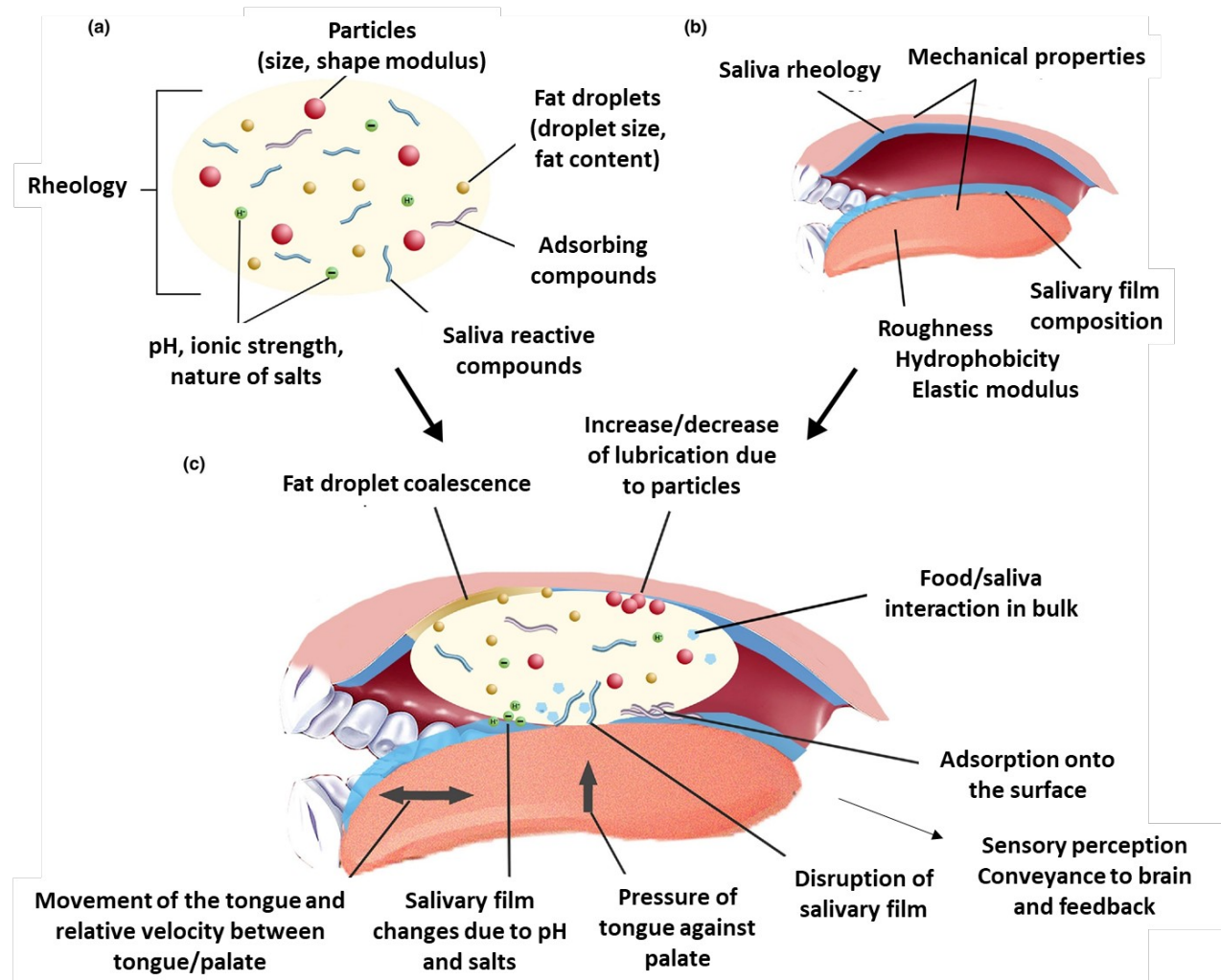


Figure 1: Graphical representation of the characteristics of (a) the food, (b) the mouth, and (c) their interaction that are important for measuring mouthfeel with tribology. Adapted from (Pradal & Stokes, 2016)

## 2.2 Quantifying mouthfeel

Guinard and Mazzucchelli (1996) state that “mouthfeel includes all of the tactile (feel) properties perceived from the time at which solid, semi-solid or liquid foods or beverages are placed in the mouth until they are swallowed” and further define residual effects of mouthfeel as after-feel, much in the

same way as after-taste refers to residual taste sensations. Before entering the mouth, food is defined by its history; i.e. composition and structure as affected by production and processing method and ingredients used. Upon entering the mouth, it is generally accepted that the mouthfeel of semi-solid and fluid foods like beverages is initially dominated by rheological properties where sensory sensations such as thickness and creaminess are perceived and, as the food is swallowed and surface interactions become more important, tribological properties begin to dominate (Chen & Stokes, 2012; Stokes, Boehm, & Baier, 2013). For solid foods the initial stage (first bite) is dominated by characteristics such as mechanical strength and fracture properties until the bolus is formed whereupon tribology becomes important (Witt & Stokes, 2015). Kokini, Kadane, & Cussler (1977) recognised that viscosity was not the only parameter necessary in order to predict sensory perceptions such as smoothness and slipperiness. Hutchings & Lillford (1988) proposed a theoretical framework for analysing the texture of food succinctly described along three axes: Time; e.g. changes in temperature and number of chews, degree of structure; bulk and particulate properties, and degree of lubrication; e.g. influence of saliva and sample moisture and fat content. The Hutchings and Lillford Breakdown Path (HL BP) provided a qualitative conceptual approach to the eating experience/perceived texture/mouthfeel using intuitive physical properties of food and additionally gave the important insight that texture exists in the brain and is therefore a psychophysical phenomenon that needs an integrative research approach combining psychology, rheology and physiology in order to be properly explained (Boehm, Yakubov, Stokes, & Baier, 2019; Hutchings & Lillford, 1988; Sarkar, Andablo-Reyes, et al., 2019). This also illustrates the importance of realising the difference between sensory properties as perceived by the brain and material properties as measured by instruments and the complexity involved in trying to directly correlate these two properties, not to mention the complexity of looking for causality in these empirical relationships (Chen, 2020). Trying to quantify mouthfeel is further complicated by the continuous transformation food undergoes after entering the mouth; i.e. structural breakdown and incorporation of

saliva causing changes in lubricating properties, meaning that determination of exactly which property; e.g. chemical, rheological, mechanical or structural, of the food-bolus at any given time correlates with a given textural sensation is an open question (Stokes et al., 2013). Recently, Boehm *et al.* (2019) proposed to adapt the HL BP into a quantitative model based on an analytical research approach, changing the nature of the model from a conceptual realm to applied, concrete recommendations. The authors stress the importance of conducting fundamental studies into foods interaction with saliva and the underlying mechanisms of lubrication of food, as well as combining several approaches (e.g. rheology, tribology) in order to provide quantifiable attributes of the breakdown of food during oral processing.

### 2.3 Surfaces – approximation to the physiology of the mouth

As previously mentioned, the choice of surface material for tribological measurements will affect the output. Two considerations are important in this aspect: getting as close to the properties of the mouth as possible and reproducibility of those conditions (Sarkar, Andablo-Reyes, et al., 2019). The need for easily accessible and cheap surface materials means that a trade-off between these two considerations will often be necessary.

In terms of surface properties, roughness ( $R_a$ ) characterised by the topography of the surface; i.e. asperities' height or well depth, width and between-distance [ $\mu\text{m}$ ] (Krzeminski, Wohlhüter, Heyer, Utz, & Hinrichs, 2012; van Stee, de Hoog, & van de Velde, 2017), hydrophobicity (wettability) measured as the contact angle ( $\theta$ ) between surface and specimen (Bongaerts, Fourtouni, & Stokes, 2007; Bongaerts, Rossetti, & Stokes, 2007), and Elastic (Young's) modulus ( $E$ ) i.e. "stiffness" [Pa] are important parameters that influence friction (Selway & Stokes, 2014).

The tongue (figure 2) is covered by four types of papillae with differing spatial distributions: filiform, fungiform, foliate and circumvallate and it is believed that the filiform papillae are responsible for



mouthfeel perception as they are the most numerous and lack taste receptors (Hanh & Frank, 2014). This renders the human tongues topography highly variable (Laguna, Bartolomé, & Moreno-Arribas, 2017). Roughness values are generally reported as ranging between 42-95  $\mu\text{m}$  (distance between asperities) with a well depth between 200-300  $\mu\text{m}$  (Godoi, Bhandari, & Prakash, 2017; Pradal & Stokes, 2016; Wang, Wang, Upadhyay, & Chen, 2019). The oral cavity is generally hydrophobic with contact angle ranging between 72-83° depending on time of day measured (Mei, White, & Busscher, 2004), but will become increasingly hydrophilic ( $\theta = 51^\circ$ ) upon addition of saliva (Sarkar, Andablo-Reyes et al., 2019). Despite this relatively rough surface (comparable to 100 grit sandpaper) the tongue does not feel rough, mainly due to its reduced Elastic modulus of approximately 2.67 kPa or 2.53 kPa for the soft palate (Cheng, Gandevia, Green, Sinkus, & Bilston, 2011). The movement of the tongue is in the range of 30 mm/s at the beginning of food intake and 5 mm/s just before swallowing, with contact pressure between 15-60 kPa between tongue and palate translating to a normal force of 0.5 N (van Stee et al., 2017); however, the tongue is capable of producing normal force ranging between 0.1-90 N (Pradal & Stokes, 2016).

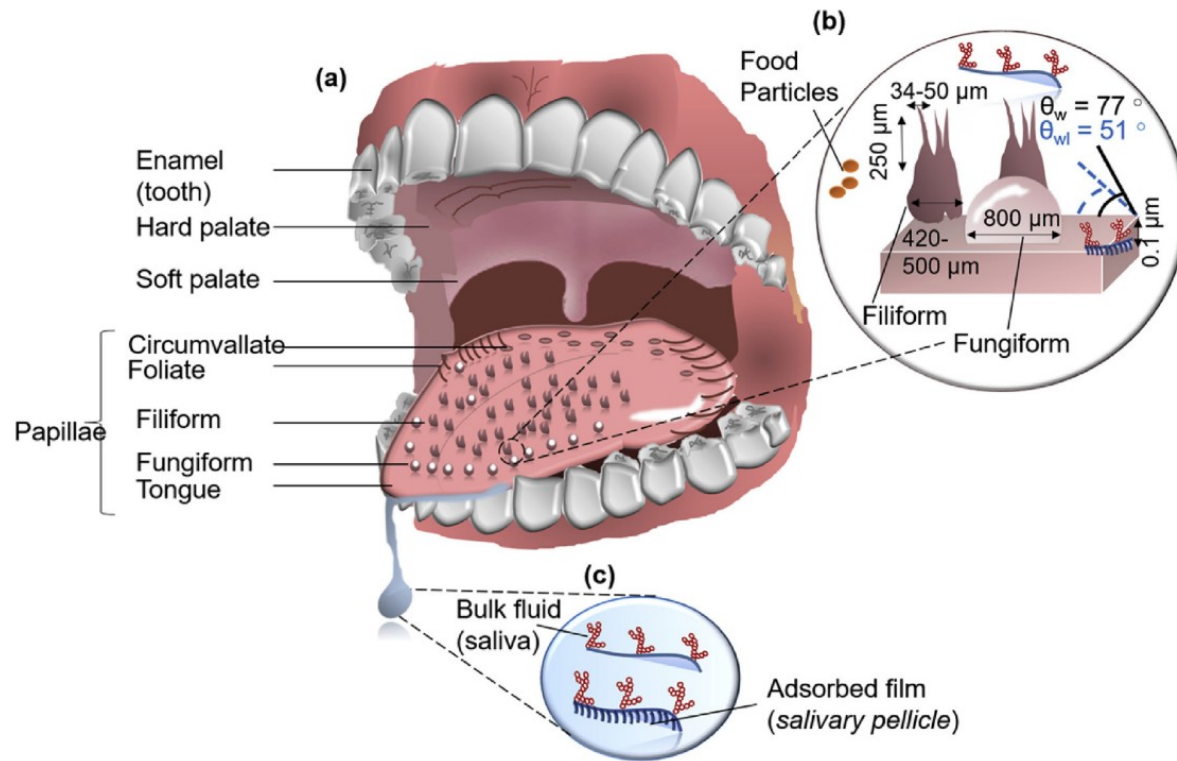


Figure 2: Graphical representation of the physiology of the mouth. (a) Schematic illustration of oral cavity highlighting the soft (tongue) and hard (tooth enamel and palate) oral surfaces with the lubricant (saliva). (b) Building blocks of soft tongue surface ( $\theta_w$  is the water contact angle,  $\theta_{wl}$  is the water contact angle upon adsorption of salivary film of nanometre scale). (c) Bulk saliva and adsorbed salivary pellicle. Reproduced with permission from Sarkar et al. (2019)

Several studies have investigated the use of biological tongues; e.g. pig's tongue (Carpenter et al., 2019; Dresselhuys, de Hoog, Cohen Stuart, & van Aken, 2008; Ranc et al., 2006), however, it is noted that variability, accessibility and issues with decomposition makes research using pig's tongue challenging. The most commonly used material for either one or both surfaces is polydimethylsiloxane (PDMS) due to its many favourable characteristics such as easily alterable hydrophobicity, roughness and surface topology (Stokes et al., 2013). PDMS is highly compliant ( $E = 0.57\text{-}3.7$  MPa) depending on cross linkage (Wang, Volinsky, & Gallant, 2014) and can relatively easily be made hydrophilic by plasma-treatment, however hydrophilicity is generally short-lasting depending on treatment (Tan, Nguyen, Chua, & Kang, 2010). The Young's modulus of PDMS is highly dependent on ratio of elastomer base to curing agent, an

aspect to be considered if production of PDMS is done in-house (Z. Wang et al., 2014). In addition, the surface of PDMS can easily be altered by casting in moulds with desired surface topography (Fitzgerald et al., 2019) and its transparent nature allows microscopical observation of lubricating behaviour in real-time (Carpenter et al., 2019). Other commonly used materials for the lower soft surface(s) include surgical tape, typically 3M Transpore Surgical Tape 1527-2, whey protein isolate (WPI), Polytetrafluoroethylene (PTFE), polyurethane and rubber; natural, foamed and styrene butadiene. For upper hard surfaces glass or steel is commonly used, but polypropylene or PDMS are also sometimes used. Carpenter *et al.* (2019) found that PDMS mimics the tongue better compared to agarose gels, however, Di Cicco *et al.* (2019) found that whey protein isolate was a better replacement for the human tongue and yielded more reproducible results compared to PDMS when applied to yoghurts.

### 2.3.1 Saliva

Saliva plays a key role during oral processing of food: it is a hydrating, lubricating, antibacterial and buffering agent, providing a medium for diffusion and/or mechanical transfer of taste-molecules to receptors, precipitation of proteins resulting in the sensation of astringency, as well as contributing significantly to enzymatic degradation and finally bolus formation and thereby safe swallowing of food (Boehm et al., 2019; Laguna & Sarkar, 2017). The composition of saliva varies significantly depending on which salivary gland it is excreted from as well as circadian rhythm, collection method, age, gender, diet, blood type and medicines (De Almeida, Grégio, Machado, De Lima, & Azevedo, 2008; Schipper, Silletti, & Vingerhoeds, 2007). In addition, it is often difficult to determine whether constituents of saliva are of human or bacterial origin and the composition of saliva will change over time as a result of contamination (Schipper et al., 2007). Generally, saliva is composed of 99% water with the remaining 1% being composed of minerals; sodium, potassium, calcium, magnesium, bicarbonate, and phosphates, nitrogenous compounds; urea and ammonia, enzymes; e.g.  $\alpha$ -amylases and lipases and immunoglobins, proteins and mucin; a glycoprotein thought to be largely but not solely responsible for the lubricating

and flow behaviour of saliva (Humphrey & Williamson, 2001; Sarkar, Xu, & Lee, 2019). Variation of any of these constituents will influence the frictional properties of saliva; either by reducing or increasing friction, by moving transition points between regimes, or by changing the slope of the curves (Sarkar, Xu, et al., 2019). Although human saliva is readily available, the use of biological samples will introduce some degree of variability, complicating interpretation and comparison of results across studies (Boehm et al., 2019). Sarkar, Xu, et al (2019) examined the use of human saliva and model saliva (i.e. artificial) and concluded that 1) although there have been advances, model saliva systems still exhibit significant differences in terms of lubricating properties, 2) out of the commercially available mucin sources, bovine submaxillary mucin is superior to pig gastric mucin and 3) more systematic research investigating model saliva systems containing mucins and polycationic additives is needed before a standardised model saliva formulation can be agreed upon (Sarkar, Xu, et al., 2019). However, recipes for synthetic saliva do exist for other purposes, e.g. *in vitro* digestion studies (Minekus et al., 2014). Although some recipes are quite different (see table 1), often the ionic composition is the same. The ionic composition of the saliva will determine the charge and pH of the solution which again will influence the solubility and configuration of the negatively charged mucins (Sarkar, Xu, et al., 2019). Further, the buffering capacity of the model saliva plays a role in stabilising the mucin proteins when mixed with food systems and adhering to surfaces (Sarkar, Xu, et al., 2019).

Compound name	Chemical formula	Upadhyay & Chen (2019)	Minekus et al. (2014)	Krop et al. (2019; Torres et al. 2019)	Laguna et al. (2017)	Cai et al. (2017)	Sarkar et al. (2019)
pH		7	7	6.8			
Sodium chloride	NaCl	0.117		0.16	1.594	0.111	1.594
Ammonium nitrate	NH <sub>4</sub> NO <sub>3</sub>			0.33			0.328
Dipotassium phosphate	K <sub>2</sub> HPO <sub>4</sub>			0.64	0.636		0.636
Potassium citrate monohydrate	KH <sub>2</sub> PO <sub>4</sub>		0.5032				
Monopotassium phosphate	KCl	0.149	1.13	0.2	0.202	1.492	0.202
Potassium citrate monohydrate	K <sub>3</sub> C <sub>6</sub> H <sub>5</sub> O <sub>7</sub> ·H <sub>2</sub> O			0.31			0.308
Uric acid	C <sub>5</sub> H <sub>3</sub> N <sub>4</sub> O <sub>3</sub> Na			0.02	0.021		0.021
Urea	H <sub>2</sub> NCONH <sub>2</sub>			0.2	0.198		0.198
Sodium lactate	C <sub>3</sub> H <sub>5</sub> O <sub>3</sub> Na			0.15			0.146
Sodium carbonate	NaHCO <sub>3</sub>	2.1	1.14			3.948	
Magnesium chloride	MgCl <sub>2</sub> (H <sub>2</sub> O) <sub>6</sub>		0.031			0.096	
Ammonium carbonate	(NH <sub>4</sub> ) <sub>2</sub> CO <sub>3</sub>		0.006				
Calcium chloride	CaCl <sub>2</sub>		0.083			0.278	
Carboxymethylcellulose	-					0.65	
Glycerin	C <sub>3</sub> H <sub>8</sub> O <sub>3</sub>					1	
Porcine gastric mucin type II	-	1.5		3	3	1.2	0-30
Alpha-amylase	-	2 g/L	75 U/mL	75 U/mL		2 g/L	

Table 1: Overview of various recipes for artificial saliva from literature

## 2.4 Stribeck curves

A Stribeck curve is the two-dimensional representation of friction coefficient as a function of (relative) sliding entrainment speed of the tribopairs, the two surfaces. The friction coefficient ( $\mu$ ) is a dimensionless number defined as the ratio between the kinetic (sliding) force ( $F_k$ ) exerted orthogonally to the normal (load) force ( $F_N$ ) (figure 3) (Blau, 2001). Assuming a constant  $F_N$  and that the kinetic force is equal to the friction force ( $F_f$ ), this linear relationship gives a quantitative measure of friction:

$$\mu = \frac{F_k}{F_N}$$

The entrainment speed ( $U$ ) is commonly presented as a dimensionless number, either the Sommerfeld number ( $\eta UR/W$ ) or, in the case of deformable surfaces, the elasto-hydrodynamic number ( $\eta UE^{1/3} R^{5/3} / W^{4/3}$ ). Load or normal force ( $W$ ), radius ( $R$ ) and Young's modulus ( $E$ ) is often considered constant so that the entrainment speed ( $U$ ) is either presented alone or scaled by the viscosity of the fluid ( $\eta$ ) in the case when viscosity changes as a function of speed, i.e. shear rate (Shewan et al., 2019; Stokes, 2012).

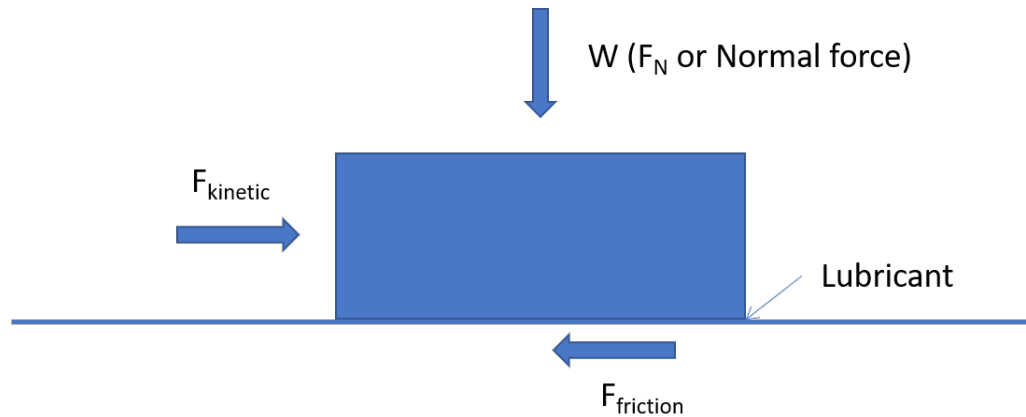


Figure 3: Graphical representation of friction showing  $F_{kinetic}$  (torque of the measuring system),  $F_N$  (the load,  $W$ ), the lubricant and the  $F_{Friction}$ .

The classic Stribeck curve (figure 4a) is often divided into three regimes depending on the film thickness between the surfaces, i.e. boundary, mixed and hydrodynamic. In the case of deformable surfaces

where the visco-elasticity of the surface influences measurements, the hydrodynamic regime is referred to as the elasto-hydrodynamic regime (Sarkar & Krop, 2019; Shewan et al., 2019; Stokes, 2012). Depending on the capabilities of the measuring system, i.e. range of speeds, a fourth regime can be included: The static regime as shown in figure 4b, which occurs at very low speeds, typically below  $10^{-6}$ - $10^{-5}$  m/s, in which movement is imperceptible (Pondicherry, Rummel, & Laeuger, 2018). This regime shows an increase in friction from 0 until a yield point signifying transition into the kinetic regime. In principle, there is no macroscopic movement in this regime and the speed depicted is due to deformation of the surfaces and the lubricant (Kieserling, Schalow, & Drusch, 2018). The boundary regime is dominated by surface properties as there is physical contact between the two surfaces' asperities and is therefore characterised by high friction coefficients, as observed by a peak or plateau. While there is fluid between the surfaces in the boundary regime, the effect is negligible compared to the impact of the two surfaces. In the mixed regime the fluid begins to be entrained between the two surfaces and thus an increase in distance and thereby contact between surface asperities is observed, resulting in decreasing friction due to thin film lubrication. In the mixed regime, effects of size of particles can be observed as the distance (D) approaches the dimensions of a given particle (Yakubov, Branfield, Bongaerts, & Stokes, 2015). In some instances "stick-slip" events are also observed in this regime, resulting in erratic behaviour and variation of the curve, the friction coefficient jumping up and down (Sanahuja et al., 2017). In the hydrodynamic regime the high speeds entrain the lubricant and generates enough lift force and hydrodynamic pressure to support the applied load and increase the distance between surfaces and is thus largely dominated by fluid dynamics. In the hydrodynamic regime the internal resistance (viscous drag) of the fluid begins to play a role leading to an increase in friction (Selway, Chan, & Stokes, 2017). It is generally assumed that the mixed and boundary regimes are highly relevant to food oral processing due to the rough and deformable nature of the tongue (Malone, Appelqvist, & Norton, 2003; Selway & Stokes, 2014).

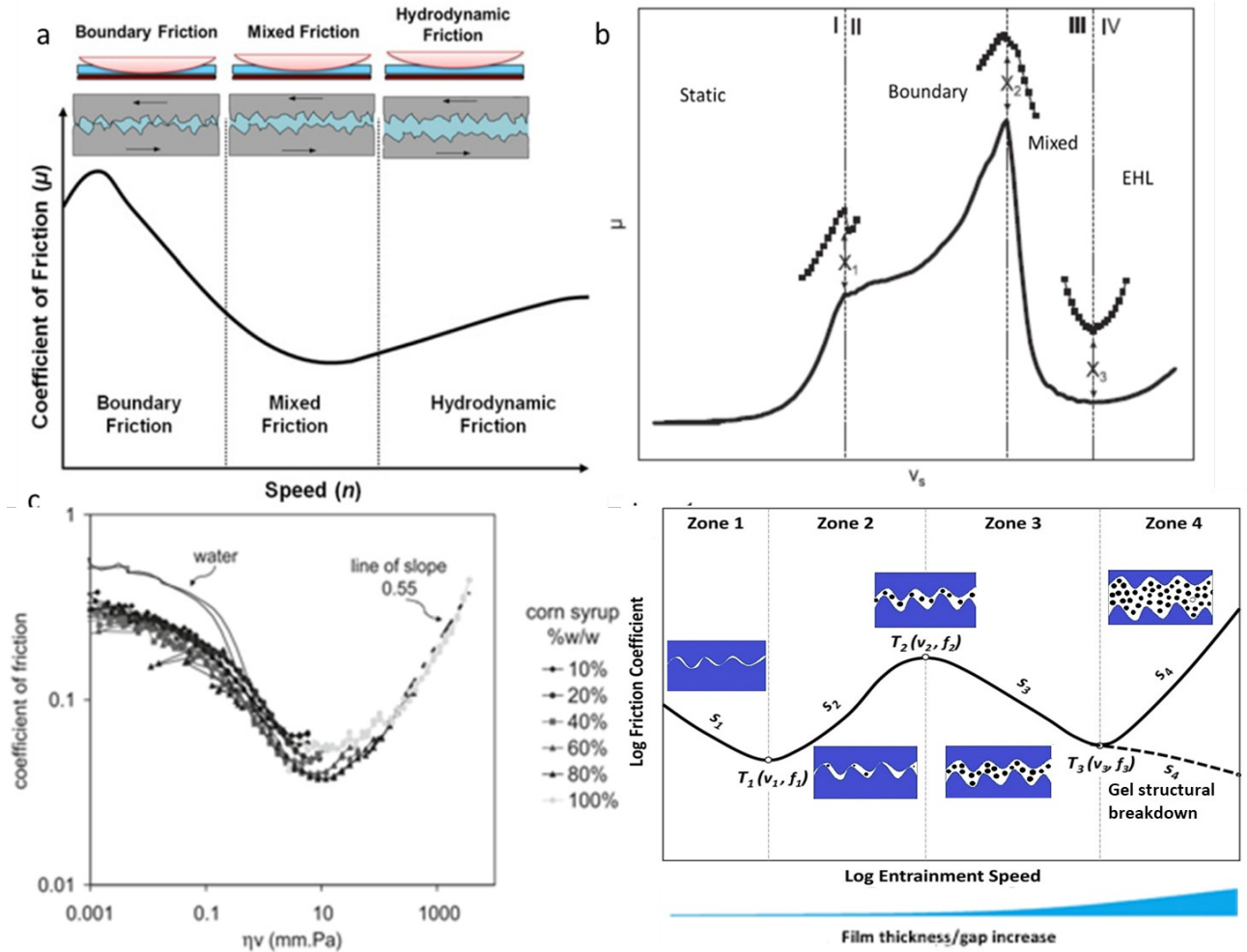


Figure 4: Four representations of Stribeck curves. (a) Classic Stribeck curve with graphical representation of gap distance ( $D$ ) and asperity interactions. Adapted from Pondicherry et al. (2018). (b) Extended Stribeck curve showing the static regime and transition points. Adapted from Kieserling, Schalow, & Drusch (2018). (c) Example of Stribeck curve obtained by collapsing several measurements on fluids with varying viscosity (entrainment speed scaled by viscosity). Adapted from Goh, Versluis, Appelqvist, & Bialek (2010). This Master curve (d) can then be approximated by fitting power law coefficients as per Bongaerts, Fourtouni and Stokes (2007). (d) Stribeck curve for a complex fluid (yoghurt) showing phase-dependent behaviour. Adapted from Nguyen, Kravchuk, Bhandari, & Prakash (2017).



## 2.5 Analysing soft tribological data – deviating from the classic Stribeck curve

The classic Stribeck curve was proposed in the early 1900s by Stribeck and colleagues working as mechanical engineers. These Stribeck curves were generated using relatively simple Newtonian lubricant and hard, non-deformable surfaces using speeds relevant to balls in ball-bearings. However, the complex and variable microstructures of food, e.g. emulsion-systems often displaying non-Newtonian behaviour, means that Stribeck curves obtained in soft-tribology using compliant surfaces will often deviate from the classic Stribeck curves (Jacobson, 2003; Rudge et al., 2019). One approach when investigating surface properties is the Master Curve Approach, where entrainment speed for several Stribeck curves for a range of Newtonian fluids with different viscosities is scaled by their respective viscosities and then collapsing these onto a single curve in a log-log coordinate system (figure 4c), thereby generating a classic Stribeck curve specific to the measuring system, lubricant and tribopairs (Bongaerts, Fourtouni, et al., 2007; Shewan et al., 2019). This Master Curve can then be approximated by fitting a set of equations involving Power law coefficients describing each part of the curve. Comparison of a Master Curve generated with hydrophilic and hydrophobic fluids with data obtained from actual complex food systems enables elucidation of the dominant phase in each regime as well as comparison with other tribopairs (Sarkar & Krop, 2019; Shewan et al., 2019).

In a slightly different approach, the entrainment speed can be scaled, i.e. multiplied, by the food system's dynamic viscosity at that shear rate if available, thereby generating a Master Curve for that food system (Joyner et al., 2014a). Care should be taken however, as the assumption that effects of viscosity are effectively “normalised” through this scaling is not necessarily valid, as viscosity and wetting behaviour of a fluid impacts the viscoelastic behaviour such as hysteresis and squeeze-out dynamics of the compliant surfaces and these effects will also alter the shape of the Stribeck curve (Selway et al., 2017).

Other approaches have also been attempted to account for the Stribeck curves obtained for complex food systems that are not easily interpretable using the terminology of classic tribology. Nguyen et al. (2017) proposed a new interpretation scheme based on data obtained for yoghurts (figure 4d). In the first zone it is assumed that initially only the fluid is entrained, and friction decreases as more and more fat globules enter the gap. With increasing speeds, a thin lubricating film is forming and the friction rises again (zone 2) until the surfaces are partly separated and the curve enters zone 3 (corresponding to the mixed regime) and finally the hydrodynamic regime (zone 4) is reached. In case of gel structure breakdown, the friction may decrease (broken line) and friction in this zone is assumed to not only be governed by viscosity but also by gel strength. A similar shape of Stribeck curve was found by Ng, Nguyen, Bhandari, & Prakash (2017). Pondicherry, Rummel and Laeuger (2018) extended the Stribeck curve to include the static regime (figure 4b), however, as the build-up of friction in this regime is assumed to be largely due to elastic and plastic deformation of the surfaces, it is still unknown whether this regime will offer insights into lubricating behaviour of food systems, but it could possibly be a valuable tool in studying the frictional properties of surfaces at nanoscales.

Different ways of interpreting Stribeck curves generated from different food systems will be further discussed in section 3.3, with an emphasis on how to obtain quantities that can be subjected to statistical analysis.

## 2.6 Rheometers with tribo-attachments (instruments)

Several rheometers have been used for tribological measurements using different measuring systems (figure 5a-c). Measuring system in this context refers to the attachment holding the surfaces; these include both commercial tribology attachments or modified rheology attachments. The measuring system setup varies, being comprised of single ball on three pins or three plates, two or three balls on plate, and ring or half-ring on plate. Rheometers used in the studies included in this review include:

MCR301, MCR302 and MCR502 (Anton Paar, Austria) and Discovery Hybrid Rheometer (DHR-3) (TA Instruments, USA).

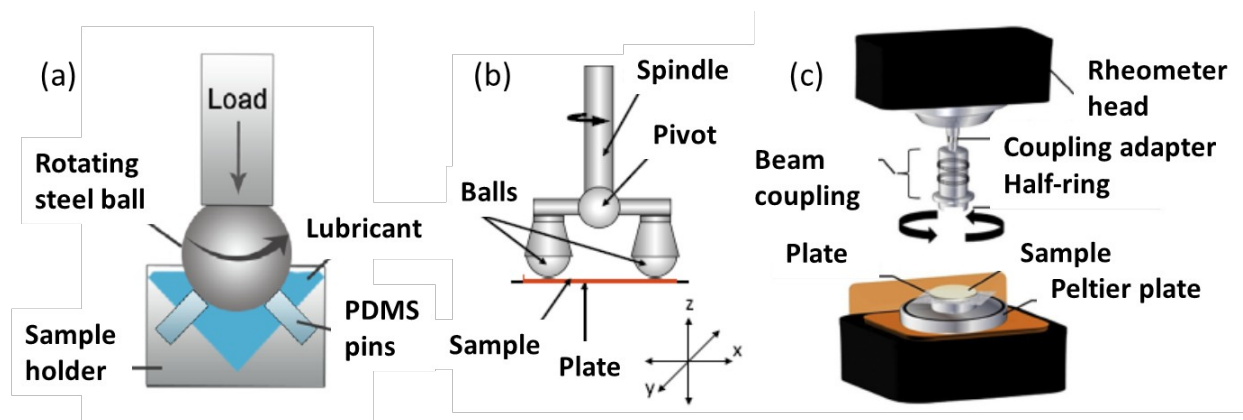


Figure 5: Main tribological attachments used in literature. (a) Ball-on-3 plates, adapted from Shewan, Pradal and Stokes (2019). (b) Double ball-on-plate, adapted from Joyner, Pernell and Daubert (2014c). (c) Half-ring-on-plate, adapted from Godoi, Bhandari and Prakash (2017)

### 2.6.1 Other tribology devices

Tribology measurements can also be undertaken using dedicated tribometers such as the MTM and the Tribolab (UMT, Bruker, Billerica USA). The MTM uses a ball-on-disk configuration that allows rotational and rolling/sliding movements as the surfaces can move independently of each other. This differs from rheometers where one surface is static. The Tribolab offers among other things a pin-on-disk setup and more complex multi-directional movement profiles that simulates the motion patterns of the tongue and can be used to study e.g. soft solid foods and boli (Campbell, Foegeding, & van de Velde, 2017; Fuhrmann, Aguayo-Mendoza, Jansen, Stieger, & Scholten, 2020; van Stee et al., 2017).

An alternative approach is to design a device or attachment in-house in order to fulfil requirements specific to the investigation. Many interesting and innovative solutions can be found in literature, ranging from custom-built attachments for rheometers (Rudge, Scholten, & Dijksman, 2020) or texture analysers (Morell, Chen, & Fizman, 2017) to devices using optical interferometry to study wetting

transitions (Martin, Clain, Buguin, & Brochard-Wyart, 2002), devices fitted with cameras or confocal microscopes for imaging-based techniques (Dresselhuis et al., 2007; Wandersman, Candelier, Debrégeas, & Prevost, 2011; Yashima et al., 2015), devices for investigating molecular organisation of soft polymer interfaces in contact (Cohen, Restagno, Poulard, & Léger, 2011) and wholly in-house built tribometers (de Wijk & Prinz, 2005).

## 3 Tribology using rheometers

### 3.1 Approaches to tribological studies

Generally, 3 approaches towards increasing understanding of mouthfeel with the aim of generating explanatory and/or predictive models can be distinguished: Conceptual, fundamental, and empirical or applied. The conceptual approach provides a theoretical framework and aims to build models that can then be tested and validated by experiments (Boehm et al., 2019; Gabriele, Spyropoulos, & Norton, 2010; Hutchings & Lillford, 1988; Kokini et al., 1977). The fundamental approach will often be applied using model fluids, e.g. concentration gradients of compound(s) of interest, and can be divided into two often overlapping categories: Methodology; development of reproducible methodologies by varying test protocols, e.g. cleaning, sample preparation, surfaces etc (Joyner, Pernell and Daubert, 2014a, 2014b, 2014c; Kieserling, Schalow and Drusch, 2018) and/or Mechanism; investigation of the underlying mechanisms of mouthfeel as described by tribological and complimentary data to generate explanatory models of mouthfeel. The latter includes e.g.:

- Investigations into the lubricating properties of saliva and its interaction with either synthetic surfaces (Bongaerts, Rossetti, et al., 2007; Carpenter et al., 2019)
- Biological materials e.g. pigs tongue (Dresselhuis et al., 2008; Ranc et al., 2006) or the food matrix (Laguna, Bartolomé, et al., 2017; Morell et al., 2017)
- The characteristics of the tribological surfaces (Dresselhuis et al., 2007; Kim, Wolf, & Baier, 2015)

- The relationship between lubricating behaviour and micro/nano-structures of the food (Garrec & Norton, 2012; Stokes, Macakova, Chojnicka-Paszun, De Kruif, & De Jongh, 2011)
- Influence of fluid viscosity and wetting on viscoelastic lubrication (Selway et al., 2017)
- Studies on the topography and physiological characteristics of the oral cavity (X. Wang et al., 2019)

The empirical (applied) approach is aimed at practical applications and strives to generate predictive models that correlate tribological (as well as chemical, rheological and physical parameters) to sensory data depending on differences in production process or composition of the given food.

## 3.2 Methodologies

A common approach when selecting measurement parameters is to choose conditions that mimic oral conditions the best as this enables elucidation of lubrication mechanisms and interpretation of results in relation to what actually happens in the mouth. There is considerable variation in measurement parameters among studies and the aim of this review is not to provide a golden standard or one-size-fits-all solution, but rather to present different methods to achieve a similar goal.

12 of the 24 studies included in table 2 below worked with dairy products (yoghurt, milk, cream cheese, custard or dairy substitutes), 7 investigated with model systems (e.g. corn syrup, mineral oil or model emulsions, as well as yoghurt), 2 worked with chocolate, and 1 each for gluten-free bread, soft drinks, and saliva. When testing a particular system e.g. reproducibility, effect of surface characteristics, and/or measuring parameters, a common practice is to use either mineral oil alone, due to its' relatively standardised lubricating properties, or a combination of demineralised water, mineral oil and yoghurt as examples of two opposites i.e hydrophilic/hydrophobic and an emulsion system exhibiting both properties.

Of the rheometers used, only two commercial producers were represented: The Modular Compact Rheometer, MCR301, MCR302 and MCR502 (Anton Paar, Austria) and the Discovery Hybrid Rheometer

(TA Instruments, USA). Steinbach, Guthrie, Smith, Lindgren, & Debon (2014) compared the use of an MCR301 with ball-on-3 plates and the MTM with single contact ball on plate on de-gassed soft drinks i.e. cola and lemon lime. And these authors reported a measured difference in friction coefficients in the boundary regime for both machines. However, the MCR301 showed an increased analytical sensitivity, as calculated by the difference in friction coefficient divided by the pooled standard deviation, by up to a factor of 200, indicating that the MCR301 with ball-on-3 plates could provide better discrimination of aqueous solutions in the lower regimes.

Table 2: Table of original papers using rheometers with tribology attachments. Abbreviations: Approaches of investigation; Mech: Mechanism, Appl: Application, Meth: Method. E.o.: Effect on/of. Rheo: Rheological properties. Tribo: Tribological properties. EPS: Exopolysaccharides.  $F_N$ : Normal force. D: Gap distance between tribopairs.  $\mu$ : Friction coefficient. T: Temperature. U: Entrainment speed; in cases where speed was not given in mm/s, this is based on own calculations. PDMS: Polydimethylsiloxane. WPI: Whey protein isolate. PTFE: Polytetrafluoroethylene. HDPE: High Density Polyethylene. SBR: Styrene butadiene rubber. NR: Natural rubber. FR: Foamed rubber. SEBS: Styrene-ethylene-butylene-styrene block co-polymer

Purpose	Food	Tribological attachment	Tribopairs		$F_N$ (N)	T (°C)	U (mm/s)	Reference
			Upper	Lower				
Saliva lubricity and cationic astringents (Mech/app)	Saliva, astringent cations	Ball-on-3 pins	Steel ball	PDMS (lab made)	6	N/A	0.01-1000, 60 mins @ 1	(Biegler, Delius, Käs Dorf, Hofmann, & Lieleg, 2016)
Modified dietary fibres e.o. structural, mechanical, sensory (Appl)	Gluten-free bread	Three-balls on sample	Steel ball	Bread	0.2	20	1	(Kiumarsi, Shahbazi, Yeganehzad, Majchrzak, & Benjamin, 2019)
Ex-vivo chocolate boluses, material properties and texture perception (Appl)	Chocolate	Ball-on-3 plates	Steel ball	PDMS (lab made)	3	40	0.02-750	(He et al., 2018)
Differentiation of two chocolate samples with identical composition and viscosity (Appl)	Chocolate	Ball-on-3 plates	Steel ball	Polyurethane	0.5	37	0.001-420	(Carvalho-da-silva et al., 2013)
Investigate discrimination by sensory compared to tribo and rheo (Appl)	Custard, starch, carrageenan, fat	Half-ring-on-plate	Steel ring	Surgical tape	2	35	0.15-100	(Godoi et al., 2017)
Development of tribological method for dairy (Meth)	Milk, cream cheeses	Half-ring-on-plate	Steel ring	Surgical tape	1, 2	35	1-600	(Nguyen et al., 2016)
Mapping in-mouth creaminess (Appl)	Yoghurt	Ball-on-3 plates	Steel ball	SBR	3	10	0.0007-667	(Sonne et al., 2014)
Surface properties ( $R_a$ ) to sensory (Meth/appl)	Demin, sunflower oil, yoghurt	Ball-on-3 plates	Steel ball	NR, FR, SBR, PTFE	3, 9	20	0.07-2000	(Krzeminski et al., 2012)
Compare whey protein isolate and PDMS for yoghurts (Meth)	Demin, sunflower oil, yoghurt	Ball-on-3 plates	Steel ball	WPI, PDMS	0.1	20	100-1, 1000-1, 200-2	(Di Cicco et al., 2019)
Whey protein phase volume, fat free yoghurts, rheology, tribo, sensory (Appl)	Fat-free yoghurts	Ball-on-3 plates	Steel ball	SBR	3	10	0.0007-667	(Laiho et al., 2017)
Gelatine, xanthan gum, carrageenan and modified starch e.o. texture of yoghurts (Mech/appl)	Yoghurt	Half-ring-on-plate	Steel ring	Surgical tape	2	35	0.008-60	(Nguyen et al., 2017)
Storage, homogenisation, pasteurisation, fat e.o. mechanical, sensory (Appl)	Milk	Double-ball-on-plate	Polypropylene	PDMS (lab made)	1	25	0,15-750	(Li, Joyner, Carter, et al., 2018)

Pasteurisation, storage and fat content e.o. rheo/tribo and astringency (Appl)	Milk	Double-ball-on-plate	Polypropylene	PDMS (lab made)	1	22	0.15-750	(Li, Joyner, Lee, et al., 2018)
Inulin, pectin, galacto-oligosacchs, beta glucan e.o. physical, rheo, tribo, sensory (Mech/appl)	Yoghurt	Half-ring-on-plate	Steel ring	Surgical tape	2	35	0.8-90	(Ng et al., 2017)
Temporal dominance sensations (TDS) and tribo (Mech/appl)	Cream cheese	Half-ring-on-plate	Steel ring	Surgical tape	2	35	0.1-600	(Ningtyas et al., 2019)
Fat e.o. tribo, rheo, structure (Mech)	Cream cheese	Half-ring-on-plate	Steel ring	Surgical tape	1, 2, 3, 5	35	0.3-300	(Ningtyas et al., 2017)
Introduction of attachment (Meth/appl)	Milk, maltodextrin, xanthan gum	Ball-on-3 plates	Steel ball	Thermoplastic elastomer	3	20	0.4-20	(Baier et al., 2009)
Tribo/rheo properties, glucone-delta-lactone or EPS cultures (Mech)	Soy yoghurt	Full ring-on-plate	Steel ring	Surgical tape	1	4	0.2-200	(Pang et al., 2019)
Demonstration reproducible results on soft drinks, comparison of MTM and MRC301 (Meth)	Soft drinks, guar gum, locust bean gum, sodium carboxymethyl cellulose	Ball-on-3 plates	Steel ball	SEBS	3	20	0.47-263	(Steinbach et al., 2014)
Method development and validation (Meth)	Demin, sunflower oil, yoghurt	Ball-on-plates/pins	Steel/glass	PTFE, PDMS, SBR	3	20	10 <sup>-6</sup> -1000	(Kieserling et al., 2018)
Validation of tribo using rheometer (Meth)	Corn syrup	Double-ball-on plate	Steel ball	Silicon	3	20	0.23-2300	(Goh et al., 2010)
Influence of measurement methodology (Meth)	Mineral oil	Double-ball-on-plate/ ball-on-3 plate	Polypropylene	PDMS, HDPE, WPI	3, 5	22	0.8-9	(Joyner et al., 2014a)
Emulsion pH, salt, homogenisation pressure e.o. friction, rheology and physics (Meth/appl)	Oil in water	Double-ball-on plate	Polypropylene	WPI	2, 1	25	10-1	(Joyner et al., 2014b)
Effect of parameter settings on F <sub>N</sub> , D, $\mu$ (Meth)	Mineral oil	Double-ball-on plate	Polypropylene	WPI, steel	1, 2, 3, 5	22	0, 1, 10 RPM	(Joyner et al., 2014c)



### *3.2.1 Tribological attachments*

The tribological attachment chosen will influence the output and hence comparison between studies using different systems, e.g. ball-on-3 plates and half-ring-on-plate, is generally not feasible. Only one of the included studies has systematically compared two different systems; comparing the use of ball-on-3 plates and double-ball-on-plate on the MCR302 (Anton Paar, Austria), Joyner et al. (2014a) found differences in the magnitude of friction coefficients of mineral oil but not in the regimes observed. This difference was attributed to the fact that the plates in the ball-on-plate system are at an angle, meaning that the small amount of oil used would have flowed to the bottom of the plates, thereby reducing the lubricating contact. More research is needed in order to quantify the potential effects of different attachments on the shape, magnitude, reproducibility, comparability, and variability of the Stribeck curves obtained.

### *3.2.2 Surfaces*

The question of whether to produce surface materials in the lab or buy commercially available surfaces comes down to a question of the aim of the study and practical considerations. While producing surface materials in-house offers control over Elastic modulus and roughness, the disadvantage is potential introduction of variability and the requirement for investigation of surface properties from batch to batch to ensure uniformity, reproducibility and accurate comparison between studies. Taking PDMS as an example, in short, the production of this polymer consists of mixing a silicone elastomer base and a curing agent, followed by degassing in vacuum to remove air bubbles and subsequent curing in an oven. The mixing ratio has profound effects on the Elastic modulus; the Elastic modulus (in MPa) can be expressed as 20 MPa/PDMS base:curing agent ratio (Wang et al., 2014). Kim et al. (2015) investigated the effects of PDMS production protocols (base:curing agent ratio, curing temperature and time, and mould finishing amongst others) on friction measurements and found that the consistency of the Stribeck curves are highly sensitive to these parameters, suggesting the implementation of a

standardised material/synthesis protocol to overcome these potential biases. Similar introduction of variability could be imagined for other in-house made polymer solutions.

The choice of surfaces used varies considerably between studies. Commonly used upper surface materials include steel (either as a ball or ring), polypropylene and glass. For the lower surfaces materials used include polydimethylsiloxane (PDMS) (either lab-made or commercial), polyurethane, surgical tape, natural rubber (NR), foamed rubber (FR), styrene butadiene rubber (SBR), polytetrafluoroethylene (PTFE), whey protein isolate (WPI), and high density poly ethylene (HDPE). Krzeminski et al. (2012) compared the use of PTFE (hard surface) and various rubbers (natural, foamed and styrene butadiene) with varying Elastic moduli (soft surfaces) and surface roughness. The harder surfaces resulted in unstable friction curve progressions and a negative correlation using Pearson's correlation coefficient from Multiple factor analysis was observed between surface roughness and friction coefficient at low speeds. This led to the conclusion that SBR was the most suitable material for discriminating between yoghurt samples measured; however, the authors did not investigate effects of wettability of the surfaces. Joyner et al. (2014a) compared the use of HDPE, WPI and PDMS using a double ball-on-plate setup and reported that WPI is the most suitable due to its' low Elastic modulus and hydrophilic nature, making it comparable to the tongue. These results are corroborated by Di Cicco et al. (2019); these authors reported a higher discriminative power of WPI compared to PDMS using a ball-on-3 plates setup when measuring several commercial yoghurt samples, showing the suitability of WPI across measuring systems. Kieserling et al. (2018) compared the use of glass or steel balls on PTFE, PDMS and SBR on a ball-on-3 plates/pins system working with demineralised water, sunflower oil, and yoghurt. Through systematic investigation of wear and reproducibility, these authors concluded that PDMS showed the least variation, however, the authors did not investigate discriminatory power when comparing similar samples, e.g. yoghurts with varying fat or protein content. The body of literature

comparing surfaces when using rheometers with tribology attachments is scarce and more research is needed.

The effects of hydrophobicity of PDMS have been studied using the MTM; adherence of hydrophobic lubricants to surfaces with low wettability, i.e. hydrophobic, results in lower friction coefficients in the boundary and mixed regimes (Bongaerts, Fourtouni, et al., 2007; Dresselhuis et al., 2007); however aqueous solutions of guar gum and xanthan gum resulted in higher friction coefficients between steel and a hydrophilic surface compared to steel and a hydrophobic surface (De Vicente, Stokes, & Spikes, 2005), indicating that the relationship between wettability and friction coefficient is not straight-forward and needs further elucidation. Besides comparing different polymers exhibiting differences in wettability, such as PDMS and WPI, another strategy is to alter the hydrophobicity of a surface material, thereby eliminating confounding variables such as Elastic modulus and surface roughness. In this regard, PDMS can be made long-term hydrophilic to varying degrees, (Hemmilä, Cauich-Rodríguez, Kreutzer, & Kallio, 2012; Shahsavan, Quinn, d'Eon, & Zhao, 2015). However, these techniques require specialised knowledge and equipment. Another possible method is the inclusion of saliva during measurements, as saliva has been shown to render surfaces hydrophilic (Macakova, Yakubov, Plunkett, & Stokes, 2011). In a study investigating the lubricating properties of whey protein microgel particles under biological conditions, Sarkar, Kanti, Gulotta, Murray, & Zhang (2017) rendered PDMS surfaces hydrophilic by plasma-treatment and reported an immediate drop in water-contact angle (from 108° to 30°) followed by a rapid recovery of hydrophobicity over 3 days before stabilisation at 63° for up to a week. By addition of a mucin layer, the contact angle of the PDMS surfaces dropped to 47° and thereby mimicked oral mucosa-coated surfaces well.

In conclusion, the choice of surfaces will depend on availability and substrate tested. WPI or PDMS reduce variability when compared to other surfaces and PDMS is a good candidate when hydrophobicity needs to be controlled or altered.

### *3.2.3 Running-in*

An often overlooked potential cause of variation in Stribeck curves is surface wear during measurements (Pradal & Stokes, 2016). Running-in refers to the initial conditioning and “smoothing” of surfaces before or during measurements until a steady-state is reached, thereby minimising effects of any differences in surface topology arising from production (Blau, 2005). Running-in presents a challenge when investigating food; consideration should be given to how often surfaces should be changed, i.e. whether it is possible to run several samples on a single surface or change with every new sample, and how to condition, i.e. prepare them for tests and reach a steady-state, the surfaces if at all before measurements. The first problem is relatively easily solved by comparing surfaces before and after a given number of runs, a run in this case meaning one sweep up or down the chosen speed range, and determining the appropriate number of runs by either statistical analysis or topographical determination. The latter does, however, require access to equipment capable of accurately characterising surfaces, such as a scanning electron microscope and atomic force microscope (Kieserling et al., 2018) or a profilometer (Arvidsson, Ringstad, Skedung, Duvefelt, & Rutland, 2017). If new surfaces are used with every new measurement (a measurement in this case can be either one single run or several consecutive runs) running in of the surfaces is necessary. Kieserling et al. (2018) conducted an in-depth investigation of running-in of PDMS, SBR and PTFE surfaces using mineral oil as lubricant. These authors did measurements comprised of 10 consecutive runs and found that a steady state was reached after approximately 5 runs, after which the obtained Stribeck curves stabilised, and the wear rate of the surfaces became defined. The first 5 runs were characterised by an undefined wear rate with high variation and a decreasing trend in friction coefficient. Additionally, the effect of multiple compressions

and between-run sample exchange were investigated; between each run, the lubricant was exchanged and the tribopairs were cleaned resulting in an increase in coefficient of variation of the Stribeck curves (Kieserling et al., 2018). Carvalho-da-silva, Damme, Taylor, Hort, & Wolf (2013) employed a similar strategy working with chocolate samples; a measurement consisted of 7-8 runs and only the last 3 were included for further analysis. A different approach was used by Steinbach et al. (2014); when investigating lubricating properties of soft drinks, a 10 minute interval at constant speed (0.47 mm/s) was employed after samples had been loaded, followed by a recording interval (single run). Goh et al. (2010) employed a similar strategy only with a 1 minute running-in period at 10 mm/s when working with corn syrup solutions. When working with chocolate samples, He et al. (2018) used a higher speed (100 mm/s) for 10 seconds. A common strategy when working with dairy products (custard, milk, cream cheese, yoghurt) is to pre-shear the samples for 1-2 mins at 1 rad/s (speed will vary depending on upper tribopair geometry) in order to ensure homogeneous distribution of sample material as well as condition the tribopairs (Godoi et al., 2017; Lee, Park, & Whitesides, 2003; Nguyen, Bhandari, & Prakash, 2016; Nguyen et al., 2017; Ningtyas, Bhandari, Bansal, & Prakash, 2017). The above examples and results carry significant implications in the case of research focused on foods with e.g. gelling properties or foods that might experience structural changes resulting in altered lubricating behaviour when subjected to shearing. If the objective of a given study is to measure lubricating properties before structural changes are induced, then reaching the steady state (defined wear rate of the surfaces) without sample exchange would prove a challenge. If, on the other hand, surfaces are pre-conditioned with a run-in period using a defined lubricant, e.g. mineral oil or a glycerol solution, then subsequent cleaning and compression of the tribopairs may introduce variation and lower reproducibility.

### *3.2.4 Entrainment speed and normal force*

Entrainment speed is generally increased or decreased, ramp-up or ramp-down, respectively, in a logarithmic fashion, so that the faster the speed is, the shorter is the time between measurement

recordings. Speed ranges used vary between studies from below 1 order of magnitude to 9 orders of magnitude (table 2) and even though most rheometers are capable of speeds down to the nanoscale, the static regime is often left unexplored. In general, the speed ranges used are chosen based on food oral processing speeds and preliminary studies to determine the best range in order to obtain the friction regimes of interest. For samples such as chocolate, yoghurt, cream cheese, custard, and corn syrup, a speed range from 0.001-0.1 up to 100-500 mm/s adequately captures the boundary, mixed and elasto-hydrodynamic regimes (Carvalho-da-silva et al., 2013; Godoi et al., 2017; Goh et al., 2010; He et al., 2018; Laiho, Williams, Poelman, Appelqvist, & Logan, 2017; Ng et al., 2017; Nguyen et al., 2017; Ningtyas et al., 2017; Ningtyas, Bhandari, Bansal, & Prakash, 2019; Pang et al., 2019; Sonne, Busch-Stockfisch, Weiss, & Hinrichs, 2014), while for more liquid samples such as milk or soft drinks, even at speeds up to 750 mm/s, the elasto-hydrodynamic regime is not observed (Baier et al., 2009; Li, Joyner, Carter, & Drake, 2018; Li, Joyner, Lee, & Drake, 2018; Nguyen et al., 2016; Steinbach et al., 2014). Although, as previously mentioned, it is generally assumed that the boundary and mixed regimes are the most relevant to measuring mouthfeel, the lack of a minimum in friction coefficient to define the beginning of the elasto-hydrodynamic regimes could cause problems in interpretation. In addition, this potentially signifies that friction at lower speed towards the static regimes could hold significant information regarding low-viscosity fluids.

Di Cicco et al. (2019) found that the narrow speed range could discriminate between non-fat and fat containing samples, but not between fat containing samples. The wide speed range resulted in higher discriminatory power, possibly due to release of fat globules during the higher shearing. Another interesting method of this study is the use of ramp-down rather than ramp-up, however, this method is not replicated in any of the other studies. As no studies have systematically investigated the influence of speed range, it is hard to make any conclusive recommendations, except to state that preliminary studies before any measurements should aim to minimise effects from speed range as well as determine

the optimum range in order to capture the relevant friction regimes. In a unique investigation, Joyner et al. (2014a), using continuous or step-wise increases in entrainment speed, found that step-wise increase resulted in the lowest variation of normal force and friction coefficient, possibly due to the system being unable to equilibrate during continuous increase. To increase comparability between studies, these authors recommend that the method of speed ramp should be reported as part of the experimental design of all tribology studies.

The effect of normal force variation has been systematically investigated on a range of attachments and surfaces. Krzeminski et al. (2012) observed that with an increase in normal force from 3 to 9 N on deformable surfaces, the overall friction is reduced. This trend is likely due to higher deformation of asperities resulting from the higher pressure (Rudge, Scholten, & Dijkman, 2020 and Urueña et al., 2018). Joyner et al. (2014c) investigated the effect of measurement parameters on normal force when using mineral oil as a lubricant and recommends proper selection of surface, rheometer base (dynamic rather than static) and dynamic normal force control (set to 100%) to reduce variation of normal force during measurements. Fluctuations in normal force during measurements can cause variability of the data, especially when using soft, deformable surfaces due to changes in contact area and normal load distribution however, Joyner et al. (2014c) notes that as normal force is part of the equation for calculating friction coefficient, small variations in normal force are mitigated. Joyner et al. (2014a) observed that friction coefficients are generally unaffected by normal force (3 and 5 N tested) when working with mineral oil. A strategy to account for any variation due to fluctuations in normal force could be to remove any data points collected when normal force was above or below a specified range (e.g.  $\pm 5\%$ ) (Joyner et al., 2014c, 2014a). Nguyen et al. (2016) tested differences in friction of dairy products (milk and cream cheese) depending on normal force (1 and 2 N) and found only small differences in friction coefficient and no differences in the regimes obtained. In a similar study, Ningtyas, Bhandari, Bansal, & Prakash (2017) investigated the effects of normal force (1, 2, 3 and 5 N) on friction

coefficients of cream cheese and found that an increase in normal force led to a decrease in friction coefficients. No clear explanations for the behaviour of non-Newtonian materials under different normal forces is currently known, but it could be due to effects on the pressure in the gap distance altering the tribological behaviour of the materials (Myant, Spikes, & Stokes, 2010). Across studies included in this review, a variety of normal forces have been applied, generally between 1-5 N. Preliminary studies should aim to pinpoint the normal force at which variation is the smallest and the highest discriminatory power is achieved. In addition, fundamental studies to investigate the effects of normal force on Newtonian and non-Newtonian materials should be undertaken. In summary, relatively narrow speed ranges are sufficient to capture the relevant friction regimes of viscous samples, e.g. chocolate spread or yoghurt. For less viscous samples, a wider speed range may capture more information. A general tendency is that friction decreases with increases in normal force. A dynamic rheometer base is preferred with dynamic normal force control set to 100%.

### *3.2.5 Temperature*

A wide range of temperatures is used in the included studies, ranging from 4-40 °C. A general trend seems to be to use higher temperatures (e.g. 35-40 °C) when investigating mechanisms of lubrication and room-temperature when defining methodologies. It is generally accepted that viscosity, density, emulsion stability, and solubility show temperature-dependent behaviour, and as such, temperature is expected to affect tribological measurements to various degrees. Although the rationale for using a specific temperature is generally justified, e.g. mimicking in-mouth conditions, the variation in temperatures used makes comparison between studies infeasible.

### *3.2.6 Cleaning of the tribopairs*

The cleaning regime used when preparing the tribopairs and tribopair holders before and between measurements will inevitably have an impact on the output, especially if even minute residues of cleaning agents, e.g. surfactants, or previous samples are left on the surfaces. As such, it is crucial that a



thorough and consistent cleaning regime is employed. In some cases, the cleaning regime is not specified, or it is unclear which cleaning agents were used (see supplementary material). Several strategies are employed and the choice of which cleaning agent (if any) to use will also depend on the specific food to be tested or the nature of the tribopairs, as well as the choice of whether or not to reuse the surface a number of times. Taking PDMS, one of the most commonly used surfaces, as an example, solvent compatibility, solvent here referring to any compounds being soluble in PDMS or able to solubilise PDMS present in either the sample or cleaning agent, has three aspects to it: (1) solubility of a given solvent in PDMS causing swelling and ensuing induction of changes to the surfaces' properties, (2) loss of solutes to PDMS causing changes in composition of the measured sample, and (3) dissolution of PDMS oligomers (potential contaminants present in the cross-linked PDMS) into the measured sample, also causing compositional changes (Lee et al., 2003 and Lee, Park and Whitesides, 2003). Going from the "lightest" to most rigorous cleaning regimes, the studies included here have employed: rinsing with deionised water and wiping with lab-wipes when working with dairy products (Nguyen et al., 2016, 2017); rinsing with isopropanol when working with mineral oil/emulsions and dairy (Joyner et al., 2014c, 2014b, 2014a; Krzeminski et al., 2012; Laiho et al., 2017; Sonne et al., 2014); rinse with ethanol when working with dairy (Di Cicco et al., 2019; Li, Joyner, Lee, et al., 2018); rinsing with detergent, followed by either a rinsing with deionised water alone (Baier et al., 2009) or using ethanol wipes (Li, Joyner, Carter, et al., 2018) when working with milk; rinsing in an acetone ultrasonic bath when working with corn syrup solutions (Goh et al., 2010); or rinsing with deionised water, followed by washing with detergent, rinsing with deionised water, followed by isopropanol, wiping with lab-wipes and drying with compressed air when working with yoghurt (Kieserling et al., 2018). The wide variety of surfaces and measurement protocols makes comparison between cleaning regimes difficult, and ultimately the choice of cleaning agents and method will be at the researchers' discretion. For the purposes of the present research, the cleaning regime of Kieserling et al. (2018) was used.

### 3.3 Data processing

Processing of the Stribeck curves obtained from tribological measurements can generally be done in two ways: semi-quantitatively by visually comparing curves for different samples in conjunction with theoretical knowledge and hypotheses, or by extraction of quantitative data for further analysis, which again can be broadly divided into two approaches. While the first is a valuable and often used tool for elucidating mechanisms of lubrication and taking into account that visual exploration of data should always be the first step in any statistical analysis if possible, visual assessment will quickly become infeasible in the context of large sample sized and multivariate data analysis. This is not to say that visual exploration of Stribeck curves is not a valid approach, but rather that generation of statistical models requires numeric data. In addition, the first approach requires in-depth knowledge of tribology and the food-matrix, while for most practical applications food scientists will be more interested in finding correlations between variables, e.g. sensory data and physical/chemical parameters. Attempts to infer statistically significant differences between samples have resulted in a few different strategies. A common pre-processing step when each data point of each run consists of several data collections is to exclude outliers above or below a certain threshold. As per good common practice, Stribeck curves are presented as mean  $\pm$  standard deviation of triplicate or more measurements for each data point and any parts of the curves of different samples not overlapping are assumed to be significantly different (Carvalho-da-silva et al., 2013; Goh et al., 2010; He et al., 2018; Joyner et al., 2014c, 2014a, 2014b; Li, Joyner, Carter, et al., 2018; Li, Joyner, Lee, et al., 2018; Ningtyas et al., 2019; Pang et al., 2019). Stribeck curves can then either be visually assessed and discussed considering complementing data or quantitatively analysed to obtain variables for further multivariate data analysis. Further extraction of numerical information generally follows two approaches; (1) comparison of friction coefficients at given set speeds, e.g. 1, 10, 100... mm/s, (Baier et al., 2009; Krzeminski et al., 2012; Laiho et al., 2017; Sonne et al., 2014; Steinbach et al., 2014) or (2) determination of  $\mu$  and  $U$  at transition points between regimes

and slopes within regimes (note that axes are generally semi-log or log-log) (Di Cicco et al., 2019; Godoi et al., 2017; Kieserling et al., 2018; Ng et al., 2017; Nguyen et al., 2016, 2017; Ningtyas et al., 2017) . The first approach seems most applicable when Stribeck curves between samples follow the same trend, i.e. transition points occur around the same speeds, or when magnitude of friction within a given regime is the object of investigation. The second approach yields information about when transitions occur depending on e.g. composition and how fast friction increases or decreases in a given regime. Taking the analysis a step further, Di Cicco et al. (2019) first extracted 8 variables from the Stribeck curves, average friction in each regime, slope in the mixed regime, and  $\mu$  and  $U$  at transition points between regimes, of 9 commercial yoghurts with varying fat-content and applied analysis of variance (ANOVA) followed by Tukey's pairwise comparison to determine which of these 8 variables best discriminated between samples. These authors then applied 2 times Principal Component Analysis (PCA) to the dataset, both the 8 variables extracted as well as the full set of measurements (9 samples x 3 runs/sample x 61 data point/run). PCA on the 8 variables in a biplot proved a valuable tool to extract information about which variables explained variation of a given sample, as well as reveal clusters of samples and correlations between variables. Similarly, PCA on the full dataset provided good separation of groups and proved a valuable tool in identifying which regimes (speed intervals) explained the largest part of the variance of the dataset.

A strategy that has so far seemingly been left unexplored is the application of calculus (e.g. area under the curve) or fitting of e.g. polynomials to bell shaped parts of the Stribeck curve.

## 4 Linking tribology and sensory

Recent reviews have examined the application of tribology as a means of explaining mouthfeel sensations and providing a link between sensory data and instrumental measurements (Sarkar & Krop, 2019; Shewan et al., 2019). Looking at the relationship between friction coefficient and sensory data

across instruments and foods, Sarkar & Krop (2019) identified three clusters based on food, sensory characteristic and friction regime: Cluster 1 contained full fat milk and yoghurt, o/w emulsions, chocolate, and cream cheese and correlations with viscosity, astringency and smoothness; cluster 2 contained low fat cream cheese, low fat yoghurt, and no fat milk and correlations with creaminess, graininess, and smoothness; cluster 3 contained emulsion-filled gels, bread, and hydrogels and correlations with roughness, fattiness, stickiness, firmness, chewiness, dryness, pastiness, slipperiness and salivating effect, with some overlap between cluster 1 and 3. Although these relationships are system-specific and have often been obtained using different sensory analysis techniques, interpretation strategies and data analyses, e.g. Pearson's correlations, PCA and Partial least squares regression (PLS), the evidence points towards tribology as a valuable tool in determining certain mouthfeel characteristics of foods (Sarkar & Krop, 2019).

Several studies have explored the link between sensory data and tribology. The link between the mouthfeel of wine, especially the attribute astringency, and instrumental measurements has been explored recently: the use of tribology has helped in elucidating some of the mechanisms responsible for this sensation, specifically the interaction between saliva and polyphenols found in wine and the correlation with friction (Laguna & Sarkar, 2017). Using a modified Texture Analyzer with stainless steel on PDMS, Stribeck curves of mixtures of whole human saliva and tannin-solutions or red wines were measured and a positive correlation was found between the friction coefficient at 0.075 mm/s and both perceived intensity of astringency and level of tannins in the samples (Brossard, Cai, Osorio, Bordeu, & Chen, 2016). In contrast, in a study using model wine systems consisting of ethanol, glycerol and oak tannins mixed with artificial saliva measured on an MTM using PDMS on PDMS, no correlation was found between presence of tannins and perceived astringency (Laguna, Sarkar, et al., 2017). A possible explanation for this could be the difference in measuring systems and experimental protocol (Laguna, Sarkar, et al., 2017), highlighting the importance of instrumental choice and setup in tribology. More

recently, in a study using the MTM and PDMS surfaces, Wang, Olarte Mantilla, Smith, Stokes, & Smyth (2020) investigated the effect of tannins and pH in wine with human saliva on level of astringency; no overall correlation could be found between astringency and friction, however, by dividing astringency into sub-qualities “rough”, “drying” and “pucker”, it was found that “drying” is driven by levels of tannins and is related to the boundary regime while “pucker” is explained by pH and rate of increase of friction. The authors conclude that explaining astringency based on interactions between saliva and astringent compounds may not be adequate and that astringency itself is multi-modal. He et al. (2018) measured Stribeck curves of expectorated chocolate boluses and found that differences in mouthcoating was reflected in the mixed regime while thickness could be correlated to the hydrodynamic regime. This is perhaps not surprising, as thickness has previously been shown to be correlated with viscosity and bulk properties (He et al., 2016; Wagoner, Çakır-Fuller, Shingleton, Drake, & Foegeding, 2019), which are the main contributors to friction in the hydrodynamic regime. These results are corroborated by Carvalho-da-silva et al. (2013), who investigated the melting and friction properties of two iso-viscous chocolate samples and found among other things, that mouthcoating and friction coefficients were negatively correlated at higher speeds. For yoghurts of various composition, e.g. differences in fat, protein, hydrocolloids and production method, it has been shown that friction coefficients at specific speeds can be successfully correlated to perceived creaminess and viscosity (Laiho et al., 2017; Sonne et al., 2014) as well as stickiness, oiliness and thickness (Ng et al., 2017; Nguyen et al., 2017). Similar results correlating creaminess/thickness to rheology/tribology data have also been found for cream cheese (Ningtyas et al., 2019). These results come together to show that correlations do exist and can be achieved by careful consideration of measuring system and protocols.

## 5 Conclusions

There has been a recent surge in studies successfully relating tribological measurements to mouthfeel of food and beverages. The need for (1) fundamental studies to determine underlying mechanisms and (2) development of standardised methods and measurement protocols to increase comparability across studies (and potentially improve correlations between sensory and tribological data) is becoming increasingly necessary. Compared to the solid knowledge base and amount of publications using the MTM, information on fundamental properties of tribological attachments on rheometers is sparse (Shewan et al., 2019). Although some of the knowledge obtained on the MTM is highly relevant and perhaps transferrable to rheometers, further investigations are needed in order to verify this assumption. Based on the above, several directions for further potential investigations have been identified:

- Effect of hydrophobicity (by incorporation of saliva, modification of surfaces, comparison of surfaces from different polymers with different wettability)
- Running-in procedures as a means to reduce variability of measurements
- Differences between tribology attachments as well as comparisons of tribometers and rheometers to determine differences in Stribeck curves and analytical sensitivity of different systems
- Influence of temperature on Stribeck curves
- Potential effects of different cleaning regimes and the chemicals used
- Potential valuable information extracted by ramp-up and ramp-down of speed, and the influence of speed range on measurements

General considerations when choosing a suitable methodology should be based on the aim of the study. More precisely, whether the measurement parameters and conditions are meant to mimic the in-mouth

conditions during oral processing as closely as possible, e.g. by using surfaces, speed ranges, normal force, and temperature etc with similar characteristics to the mouth, or whether the focus should be on capturing as much data as possible, e.g. wide speed range, with as high a discriminating power and reproducibility as possible, e.g. for correlations with sensory and compositional data etc. This trade-off will influence the possible interpretations of the Stribeck curve, and the data obtained will reflect these considerations. Naturally, the data analysis and information extraction should be tailored to the specific aim, whether it be explanatory or predictive power. Figure 6 gives a graphical representation of parameters to consider at each step of planning a tribological study.

For better comparability between studies, it is recommended to:

- Conduct preliminary studies to determine best speed range, running-in procedure, cleaning regime
- Report in detail on production method of surface polymers, cleaning regime, temperature, running-in procedure, and method of speed ramp
- Use surfaces with standardised characteristics and in the case of in-house made surfaces report surface roughness, wettability and Elastic modulus
- Gather as much data as possible on the samples to provide high statistical power, and potentially conduct multivariate data analysis on friction data alone (to eliminate redundant variables and identify relevant variables/friction regimes) and in conjunction with other data collected

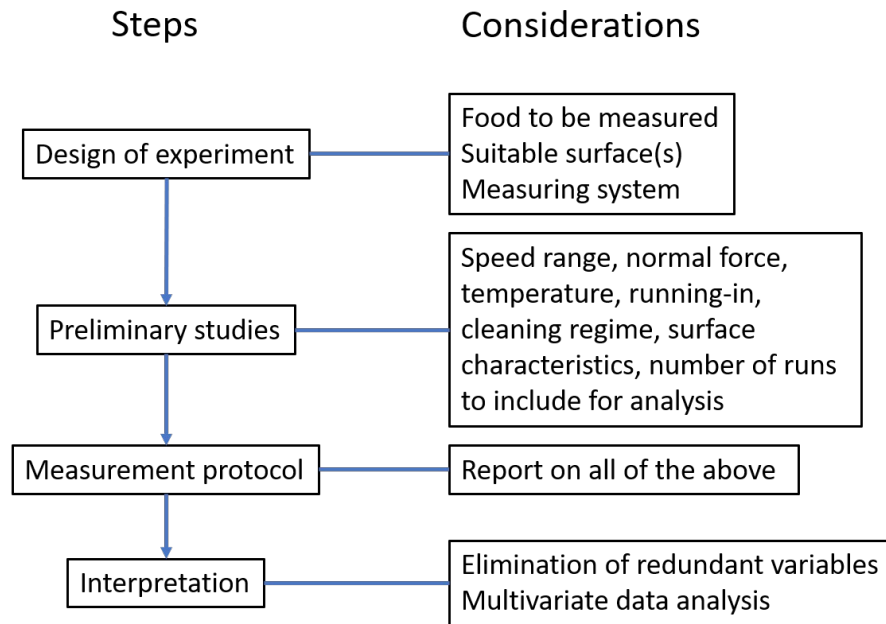


Figure 6: Graphical representation of suggested flow chart highlighting important steps and considerations at each step.



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# Chapter II Development of a tribology method and correlation with sensory data from an online database

## 1 Introduction

The mouthfeel of any beverage is an important indicator of acceptance and liking by consumers (Guinard & Mazzucchelli, 1996), and beer is no exception. Current trends in the beer market are the continuing rise and increasing market share of craft breweries (BA, 2019; ICBI, 2018) with an accompanying demand for beers with more complex sensory attributes than the traditional lager beer (Gabrielyan et al., 2014; Thurnell-Read, 2018), as well as increasing interest in non-alcoholic beers (NAB) and low-alcoholic beers (LAB) (Bellut & Arendt, 2019). The choice of beer by consumers is influenced by a number of factors, ranging from extrinsic factors such as brand, price, alcohol content; sociodemographic and cultural background; biological, genetic, and psychological characteristics; the context and environment of consumption; as well as product-intrinsic factors such as sensory attributes of the beer (Betancur et al., 2020). In the context of NABs, mouthfeel is often described as being deficient, especially in NABs produced by physical dealcoholisation processes (Krebs et al., 2019; Malfliet et al., 2009; Müller et al., 2017; Schmelzle et al., 2013). The mouthfeel of beer is attributed to a diverse number of beer constituents and can be modified by changing ingredients and processing method. The content of ethanol has a significant impact on the perception of mouthfeel, as exemplified by research on NABLABS (Krebs et al., 2019; Ramsey et al., 2018). In addition, polyphenols (Goiris et al., 2014; Wannemacher et al., 2018), dextrans (Rübsam et al., 2013), chloride ions, glycerol, beta-glucan (Langstaff et al., 1991a), proteins (Langstaff & Lewis, 1993; Steiner et al., 2012), and arabinoxylan (Langenaeken et al., 2020) are key contributors to mouthfeel of the final product. The concentration of these compounds can be altered by, e.g. brewing with adjuncts (Bellut et al., 2019) or altering the mashing profile (Krebs et al., 2020), but will also be altered by the dealcoholisation process (Krebs et al.,

2019; Müller et al., 2017). In the search for innovative products using novel ingredients or production methods, rapid screening protocols of prototypes are often required to circumvent costly and time-consuming sensory trials. Recent years has seen a rise in the use of various experimental approaches to quantifying mouthfeel using a combination of techniques often including tribology in combination with rheology and chemical characterisation (Fox et al., 2020; Sarkar & Krop, 2019; Shewan et al., 2019). Mouthfeel is a complex sensory percept elicited by physical, tactile sensations as governed by the interaction between the food or beverage and the surfaces found in the oral cavity (Sarkar et al., 2019). In the early 1990s, pioneering work (Langstaff & Lewis, 1993; Langstaff et al., 1991a, 1991b) explored the mouthfeel of beer and proposed a modification of the position of mouthfeel, previously placed as a subcategory under the headings aroma and taste (Meilgaard et al., 1979), in a category of its own (Langstaff et al., 1991b). Furthermore, mouthfeel was divided into three main classes: 1. Carbonation; sting, bubble size, foam volume and total carbon dioxide 2. Fullness; density and viscosity, and 3. Afterfeel; oily mouthcoating, astringency and stickiness. A further modification was proposed more recently, with an emphasis on clearer communication with consumers (Schmelzle, 2009): 1. Mouthfeel; tingly, warming, astringent, pungent, 2. Body; density, viscosity and 3. Foam; volume and structure.

The purpose of the present research is to 1. Determine the chemical composition as well as pH, total titratable acids (TTA) and extract of 10 beers, 2. Develop a method for measuring frictional parameters of beers and NABs, and 3. Analyse the tribological characteristics of the selected beers.

## 2 Methods

A shotgun-approach was used, whereby as many variables as possible were extracted from the friction curves and then the number of variables was reduced by various dimension reduction techniques: PCA, clustering and Spearman's correlation. Secondly, sensory data from an online database on these 10 beers were collected and used to 1) compile keywords chosen by consumers to describe mouthfeel of

beer and 2) conduct multivariate data analysis to examine correlations between physical and chemical characteristics and mouthfeel of the beers.

## 2.1 Beer samples

A range of different styles of beers was selected based on availability of a non-alcoholic counterpart aimed at mimicking as closely as possible the alcoholic version. Five different beers; Heineken (lager), Krombacher Pilsner, Leffe blond (Belgian lager), Hoegaarden witbier (wheat beer), and Paulaner hefeweiss (wheat beer) and their non-alcoholic version were chosen representing different dealcoholisation processes. Samples were prepared by degassing for 10 mins in an ultrasonic bath, followed by filtering through Whatman™ filter paper grade 1 (Merck, Darmstadt, Germany). Filtering was done to remove any residues, e.g. lees, from the beers in order to be able to perform further analysis. While this will inevitably have an impact on the mouthfeel of the samples, it is assumed that this is negligible.

## 2.2 Characterisation of beers

### 2.2.1 *Sugars and acids*

Sugar contents of the beers were determined by high performance liquid chromatography (HPLC) Agilent 1260 Infinity (Agilent Technologies, Santa Clara CA, U.S.A.) equipped with a refractive index detector (RID) and a Sugar-Pak I 10 mm, 6.5 mm by 300 mm column (Waters, Milford MA, U.S.A.) with 0.1 mM of Ca-EDTA as the mobile phase and a flow rate of 0.5 mL/min at 80 °C. Organic acids were quantified by HPLC (Waters 2690 Separations Module, Waters, Milford MA, U.S.A.) with diode array detector (Agilent Technologies, Santa Clara CA, U.S.A.) with 5mM (DAD) and a Hi-Plex H 8 mm, 7.7 mm by 300 mm column with 5 mM H<sub>2</sub>SO<sub>4</sub> as the mobile phase and a flow rate of 0.5 mL/min at 60 °C.

### 2.2.2 pH, TTA, extract and ethanol content

Total titratable acidity and pH was measured on an EasyPlus™ Titration (Mettler Toledo, OH, USA).

Extract and alcohol content were measured using a density meter DMA 4500M with an Alcolyzer Beer ME (Anton Paar GmbH, Graz, Austria).

### 2.2.3 Viscosity

Dynamic viscosity measurements were performed using a Haake falling ball viscometer Type C (Thermo Fisher Scientific, MA, USA) according to MEBAK method 2.25.1.

## 2.3 Tribology

Tribological measurements were carried out using an MCR301 using the BC-12.7 ball-on-3-pins tribology attachment (Anton Paar GmbH, Graz, Austria). Tribopairs, glass balls and PolyDiMethyl Siloxane (PDMS) pins, were supplied by Anton Paar (Pondicherry et al., 2018). The choice of surface, as well as cleaning regime and test configuration, was based on Kieserling et al. (2018). Briefly, new tribopairs were used for each test; a test consisted of a run-in period and three consecutive measurements of friction curves. Before each test, the pin-holder and ball-holder were washed gently using a dilute detergent solution, rinsed thoroughly with demineralised water and wiped with 70% ethanol before being dried using compressed air. Tribopairs were rinsed twice in acetone and dried with lab-wipes before being dried using compressed air and subjected to a final visual examination.

The following measuring system parameters were used: Trurate™ was set at 80%, normal force dynamic of 50% was chosen, and range limitation was set at 150 mN\*m. All measurements and run-in were performed at a normal force of 3 N and a temperature of 20 °C. Measuring system inertia was calibrated, and normal force was set to zero before each test. The glass ball was lowered slowly until the desired normal force was reached and then held for 2 mins for equilibration. A run-in sequence consisting of 5 speed steps logarithmically increasing from  $10^{-4}$ - $10^0$  m/s (5 mins at each speed) was

employed, followed by a resting period of 2 mins. For each sample, three consecutive measurements of friction curves were performed using stepwise, logarithmic increases in speed (from  $10^{-8}$ - $10^0$  m/s), recording 80 data points with logarithmically decreasing ramps in data recording intervals (from 10-1 s). Between each measurement, a rest of 90 s was employed. The two last runs of each test were used for further analysis, and each sample was measured in independent triplicates.

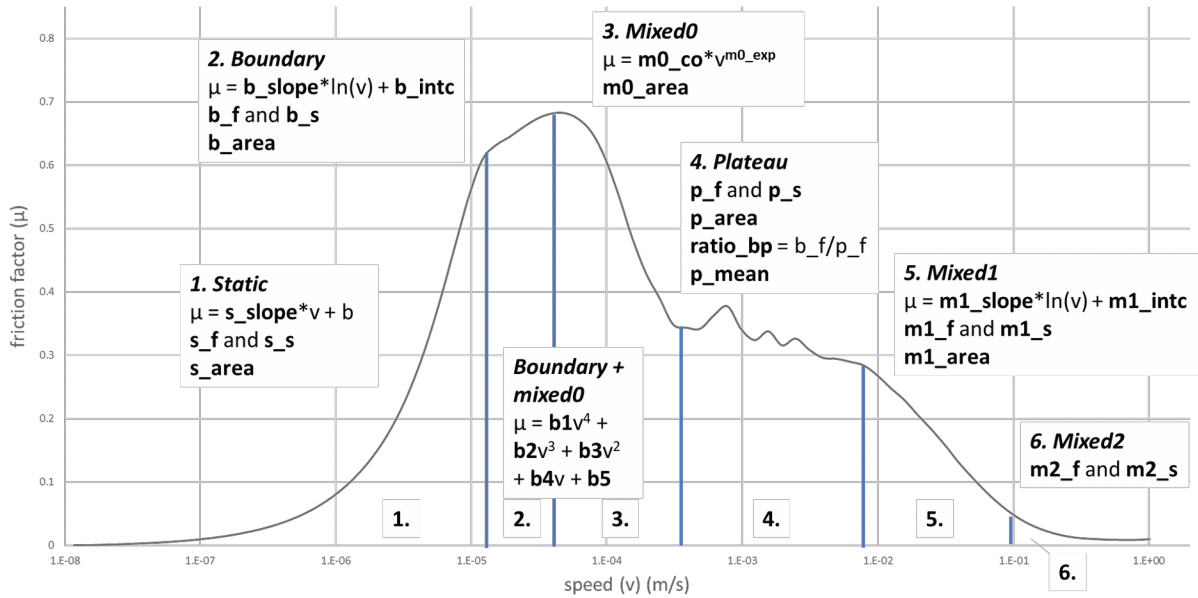


Figure 7: Overview of variables extracted from the friction curves and the friction regimes identified for the beers.  $s\_slope$ : linear slope of the static regime;  $s\_f$  and  $s\_s$ : friction and speed at the end of static regime;  $s\_area$ : area under the curve of static regime;  $b\_slope$  and  $b\_intc$ : slope and intercept of logarithmic trendline of boundary regime;  $b\_f$  and  $b\_s$ : friction and speed at the end of boundary regime;  $b\_area$ : area under the curve of boundary regime;  $b1-5$ : parameters of 4th-degree polynomial fitted to boundary and mixed0;  $m0\_co$  and  $m0\_exp$ : coefficient and exponent of power law fitted trendline of the mixed0 regime;  $m0\_area$ : area under the curve of mixed0;  $p\_f$  and  $p\_s$ : friction and speed at the start of plateau;  $p\_area$ : area under the curve of plateau;  $ratio\_bp = b\_f / p\_f$ ;  $p\_mean$ : mean friction in plateau;  $m1\_slope$  and  $m1\_intc$ : slope and intercept of logarithmic trendline of mixed1;  $m1\_f$  and  $m1\_s$ : friction and speed at the beginning of mixed1;  $m1\_area$ : area under curve of mixed1;  $m2\_f$  and  $m2\_s$ : friction and speed at the beginning of mixed2.

Data processing and extraction of relevant variables were carried out in excel. Figure 7 describes which parameters for each regime were extracted. These include the friction factor and speed at transition points between regimes; parameters obtained by fitting a trendline to each regime as appropriate, i.e. linear, logarithmic, polynomial, exponential or power-law; the area under the curve of each regime, obtained by manual integration of the curves; as well as the ratio between the maximum of the boundary regime and onset of the plateau after the first mixed regime period.

The obtained variables were then subjected to PCA (supplementary figure 1). After inspection of the screeplot (supplementary figure 2) PCs 1 and 2 were retained, accounting for 52.21% and 15.52% of variation, respectively. Variables with high quality of representation ( $\text{Cos}^2$ ) on PCs 1 and 2 (supplementary figure 3) were considered for further use and subjected to cluster analysis. Based on clustering (supplementary figure 4) using the varclus function (hierarchical clustering, complete linkage) based on the similarity matrix of Hoeffding's D statistic to account for monotonic and non-monotonic relationships from Hmisc package (Harrell Jr & Others, 2020), variables were grouped and, after inspecting the correlation matrix (Spearman's rho) of tribology variables alone as well as tribology variables with sensory data (supplementary figures 5 and 6), variables describing the same quality and exhibiting the same pattern of correlations were further grouped for ease of interpretation and reduction of redundancy.

## 2.4 Sensory data

Sensory data was collected from [www.ratebeer.com](http://www.ratebeer.com), a website that provides a free platform for submitting and viewing reviews of beers. For each beer between 28-191 reviews were used depending on availability. This number was chosen to avoid a disparate number of observations between beers as some of the beers (especially the NABs) had a relatively low number of reviews (~30). The selection

criteria for the reviews were the number of reviews submitted by the reviewer. This choice was made on the assumption that reviewers with a high number of reviews were more likely to 1) have a vested interest in providing accurate and meaningful reviews, i.e. to maintain their reputation and be respected by their fellow beer-reviewers and 2) be competent beer-tasters with experience and ability to accurately assess and describe mouthfeel (Giacalone et al., 2016; Van Doorn et al., 2020).

The written reviews were converted to a text corpus and uncluttered by removing common English stop-words (e.g. and, or, if etc.), numbers, white space, punctuation, and words were lemmatised (Welbers et al., 2017). Lemmatisation is the process of grouping together inflected words (e.g. "pours", "pouring" and "poured" are reduced to the root-word, or lemma, "pour") according to a dictionary of English. This process is not always perfect and sometimes leaves words with the same lemma in the corpus as can be seen in table 3 below (e.g. "sour" and "sourness"). The reviews were then converted into a DocumentTermMatrix (DTM): a matrix where rows are reviews and columns are words, and each cell contains the frequency (e.g. 1, 2, 3 etc.) of a given word in that review. A list of 32 terms commonly used to describe or associated with mouthfeel of beer was created based on literature findings ("mouthfeel", "mouth", "feel", "body", "bodied", "aftertaste", "taste", "water", "watery", "smooth", "creamy", "palate", "light", "heavy", "thin", "thick", "crisp", "clean", "clear", "foamy", "carbonation", "head", "full", "flat", "warm", "round", "oily", "astringent", "astringency", "texture", "foam", "carbonate") (Guinard & Mazzucchelli, 1996; Langstaff et al., 1991b; Schmelzle, 2009). A function designed to find associated words, the 'findAssocs' function from tm-package (Feinerer & Hornik, 2019), searched the DTM for mouthfeel associated terms, resulting in a total of 2202 descriptors. After filtering for words with a frequency higher than 10, a total of 300 words remained. These were manually cropped to 49 words by removing words not holding any association with mouthfeel. This list was then checked for correlation to mouthfeel by only retaining words significantly correlated ( $p < 0.05$ ) to words with "neutral" mouthfeel connotations, e.g. "body", "mouth", "mouthfeel", "bodied", "aftertaste", "tongue",



"palate", as well as the numeric mouthfeel rating and further reduced to 23 distinct terms by removing ambiguous terms, e.g. "finish", "palate", "mouthfeel" etc.. These terms were clustered using the varclus function, based on Hclust.method, from Hmisc package (Harrell Jr & Others, 2020) to produce a dendrogram (figure 14).

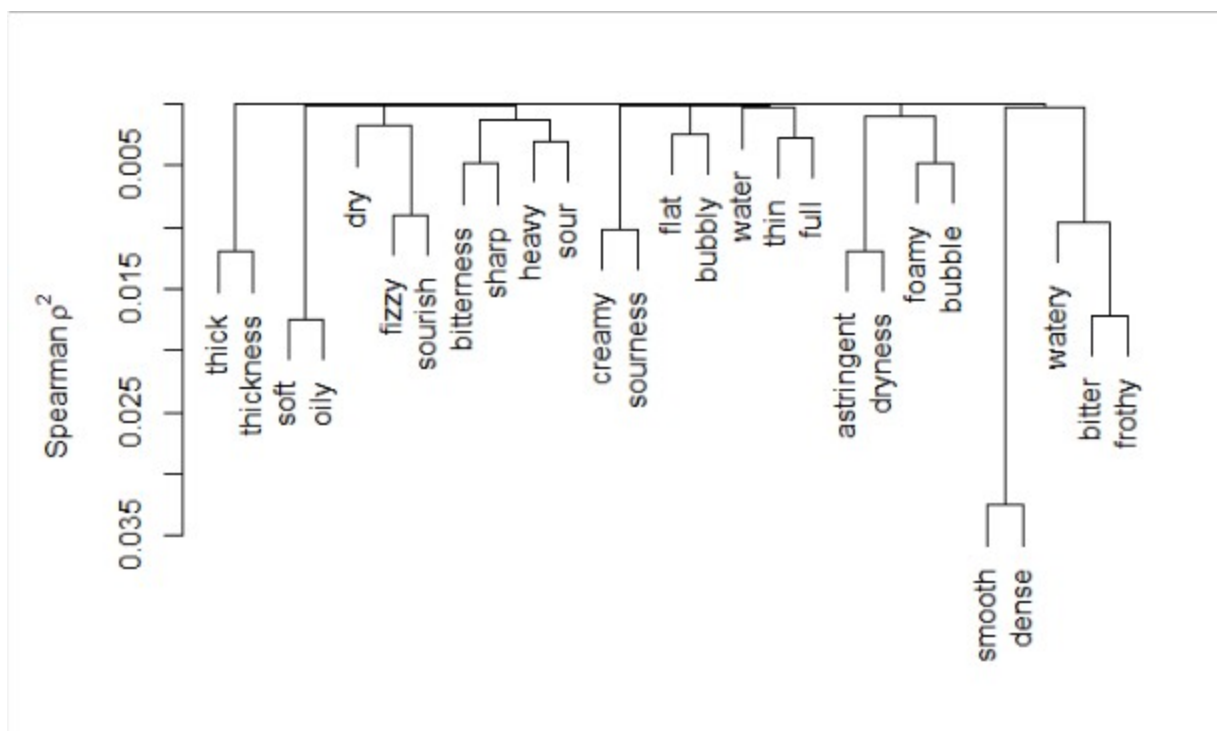


Figure 8: Dendrogram displaying similarities (Spearman's  $\rho^2$ ) between mouthfeel terms.

Based on these relationships and at the discretion of the authors, the terms were grouped into 7 overall mouthfeel attributes: "watery", "smooth", "bitter", "thick", "foam", "astringent", and "sour" (table 3). Perceived bitterness and sourness and related terms were included even though they technically fall under the category of taste, as consumers often use these words to describe the sensation of astringency (Vidal et al., 2015).

Water	0.25	Smooth	0.23	Bitter	0.29	Thick	0.12	Foam	0.15	Astringent	0.03	Sour	0.08
y		h											
Thin	0.11	Smooth	0.07	Bitter	0.16	Dense	0.02	Foamy	0.03	Astringent	0.005	Sour	0.04
Watery	0.11	Creamy	0.09	Bitterness	0.13	Full	0.02	Bubbly	0.02	Dry	0.02	Sourness	0.01

Water	0.02	Soft	0.07	Heavy	0.02	Bubble	0.02	Dryness	0.01	Sourish	0.01
Flat	0.01	Oily	0.01	Thick	0.05	Frothy	0.05			Sharp	0.01
				Thickness	0.00	Fizzy	0.03				
					1						

Table 3: Relevant mouthfeel descriptors and their frequencies.

## 2.5 Statistics and R-packages

Statistical analysis was done using R-studio (RStudioTeam, 2020). For text-mining, the package tm (Feinerer & Hornik, 2019) was used. For Principal Component Analysis (PCA), the packages FactoMineR (Lê et al., 2008) and factoextra (Kassambra & Mundt, 2020) were used. Evaluation, processing, and extraction of tribological parameters (min/max, mean, area under curve, trendline fitting) was done using Excel. Trendlines; linear, polynomial, power, logarithmic or exponential, were fitted manually to the friction curves by determining beginnings and ends of regimes and excluding values at either end until an  $R^2 > 0.95$  was achieved. Analysis of variance was, where relevant, followed by Tukey's test in R using the agricolae package (de Mendiburu, 2020). For all analyses, where relevant, a significance level of  $p < 0.05$  was applied.

## 3 Results and discussion

### 3.1 Chemical and physical data

Table 5 summarises the chemical and rheological data on the 10 beers. Information on the exact method of de-alcoholisation is not freely available for all the beers included but based on the chemical data, some educated guesses can be made. The glycerol content of Heineken NA is low, indicating that this beer was produced by limited fermentation. According to the Heineken website, it is double-brewed followed by vacuum-distillation and blending with natural flavourings. All the NABs have  $<0.1$  % ABV except for Paulaner NA with 0.4 % ABV, indicating that this NAB was possibly made by blending a de-alcoholised beer with a normally brewed beer. The low content of fermentable sugars in Leffe NA indicates that this was fully fermented and then de-alcoholised, while both Krombacher NA and

Hoegaarden NA were possibly blended after being fully fermented and de-alcoholised. The beers are very different in viscosity, ranging from 1.2 for Heineken NA to 1.7 for Leffe NA. Interestingly, no correlation was found between pH and TTA or between TTA and citric acid or lactic acid concentration (table 4), which could be related to the buffering capacity, either naturally occurring or altered, of the worts/beers (Peyer et al., 2017).

Table 4: Correlation matrix (Pearson *r*) of chemical data. tta: Total Titratable Acids; rex: Real extract; oex: Original extract; abv: Alcohol by volume (%); suc.mal: Sucrose/maltose; glu: Glucose; fru: Fructose; man: Mannitol; cac: Citric acid; lac: Lactic acid; glyc: Glycerol. Numbers in bold are statistically significant ( $p < 0.05$ ).

	pH	tta	rex	oex	abv	suc.mal	glu	fru	man	cac	lac	glyc
pH	1.00	0.03	<b>-0.53</b>	<b>-0.58</b>	-0.20	0.16	-0.12	-0.15	-0.24	-0.20	<b>0.40</b>	<b>-0.49</b>
	tta	1.00	-0.18	0.24	0.27	-0.16	0.12	0.19	<b>0.53</b>	0.01	0.02	0.04
		rex	1.00	-0.31	<b>-0.70</b>	0.36	<b>0.73</b>	<b>0.63</b>	-0.08	-0.13	0.06	0.12
			oex	1.00	<b>0.90</b>	<b>-0.54</b>	<b>-0.43</b>	-0.29	<b>0.47</b>	0.31	<b>-0.48</b>	<b>0.51</b>
				abv	1.00	<b>-0.57</b>	<b>-0.66</b>	<b>-0.51</b>	<b>0.39</b>	0.29	<b>-0.39</b>	0.32
					suc.ma							
						1.00	0.14	-0.13	<b>-0.69</b>	<b>-0.79</b>	<b>0.66</b>	-0.16
					I							
						glu	1.00	<b>0.96</b>	0.06	-0.07	0.08	0.09
							fru	1.00	0.27	0.15	-0.10	0.12
								man	1.00	<b>0.59</b>	<b>-0.46</b>	0.11
									cac	1.00	<b>-0.73</b>	-0.24
										lac	1.00	<b>-0.36</b>
											glyc	1.00

Beer	pH	TTA	Lactic acid	Citric acid	Sucrose/ maltose	Glucose	Fructose	Mannitol	Glycerol	% ABV	Real extract	Original extract	Viscosity
Heineken	4.6±0.0 6	1.6±0.03	0.0±0.0	0.3±0.0	0.7±0.0	0.0±0.0	0.1±0.01	0.8±0.01	0.4±0.0	4.2±0.0	3.1±0.0	9.5±0.0	1.3±0.0
Heineken NA	4.6±0.0 4	1.9±0.02	0.4±0.0	0.3±0.0	10.6±0.0 1	2.2±0.0	1.1±0.01	0.8±0.0	0.1±0.0	0.1±0.0 1	5.2±0.0	5.3±0.01	1.2±0.0
Hoegaarden	4.4±0.0 1	1.9±0.03	0.4±0.0	0.2±0.01	0.7±0.0	0.0±0.0	0.1±0.0	1.1±0.0	0.4±0.0	5.3±0.0	4.3±0.0	12.4±0	1.4±0.01
Hoegaarden NA	4.4±0.0 6	2.4±0.1	0.2±0.0	0.2±0.01	2.2±0.01	14.6±0.02	13.3±0.02	1.3±0.0	0.4±0.0 1	0.0±0.0 1	7.6±0.0	7.7±0.01	1.3±0.0
Krombacher	4.4±0.0 1	2.2±0.08	0.1±0.0	0.3±0.0	1.3±0.0	0.0±0.0	0.1±0.0	0.8±0.0	0.4±0.0	5.4±0.0	3.8±0.0	12.0±0.01	1.4±0.0
Krombacher NA	4.4±0.0 7	1.8±0.03	0.4±0.01	0.0±0.0	25.9±0.0 2	6.2±0.01	2.3±0.0	0.1±0.0	0.4±0.0	0.1±0.0 1	7.8±0.0	7.9±0.01	1.4±0.0
Leffe	4.1±0.0 2	2.1±0.15	0.0±0.0	0.3±0.0	2±0.0	2.0±0.01	1.9±0.01	1.4±0.0	0.4±0.0	6.9±0.0 1	6.0±0.0	16.2±0.01	1.5±0.03
Leffe NA	4.1±0.0 5	1.5±0.05	0.0±0.0	0.3±0.04	0.3±0.0	6.7±0.01	6.0±0.0	1.0±0.0	0.4±0.0	0.0±0.0 2	9.6±0.01	9.7±0.04	1.7±0.01
Paulaner	4.4±0.0 1	2.4±0.06	0.1±0.0	0.2±0.01	0.9±0.0	0.0±0.02	0.1±0.01	1.3±0.0	0.4±0.0	5.6±0.0 2	4.5±0.01	13±0.02	1.6±0.01
Paulaner NA	4.4±0.0 1	2±0.05	0.2±0.0	0.1±0.0	18.7±0.0 1	2.7±0.0	0.7±0.0	1±0.01	0.4±0.0	0.4±0.0 1	7.0±0.0	7.7±0.01	1.5±0.01

Table 5: Chemical data and viscosity of the beers. TTA is presented as mL NaOH/10 mL sample; lactic acid, citric acid, sucrose/maltose, glucose, fructose, mannitol and glycerol are presented in g/L; % ABV is per cent alcohol by volume; extracts are presented in ° Plato; viscosity (dynamic) is presented in mPa\*s.

## 3.2 Tribology

For the static regime, the intercept was excluded from analysis as this parameter is zero in all cases (table 6); for the plateau, no trendline was fitted because of the large fluctuations in friction leading to low  $R^2$ ; for the mixed regime, no trendline was fitted because of too few data points. A large number of variables were eventually grouped under mean friction of the plateau ( $p\_meanf$ ). Additionally,  $m2\_s$  was retained despite a low  $\cos^2$  based on its' correlation with astringency.

Table 6: Overview of variables retained from friction curves.

Retained	Overlap	Dropped (low Cos2)
s_slope	s_s	b_slope
b_intc	b_area	b1-4
p_meanf	p_area, m1_f,	m0_exp
	s_area, s_f, b_f,	m0_co
	b_s, ratio_bp,	m1_s
	b5, m0_area,	m1_area
	p_f, p_s	
m1_slop	m1_intc	
e		
m2_s	m2_f	

A general trend is that NABs have lower overall friction coefficients than their alcoholic counterpart. The wheat-beers, normal and NA all exhibited high lubricating properties while Krombacher, normal and NA, Heineken, normal and NA, and Leffe, normal, have high friction coefficients. Leffe NA positions itself in the middle and as such confirms itself as an outlier according to tribological measurements as well as sensory data (section 3.3). The tribological profile of the beers differs from that of the classic Stribeck curve in several ways. The classic Stribeck curve is commonly divided into boundary, mixed and elasto-hydrodynamic regimes (Sarkar et al., 2019). The present curves include the static regime as well as a mixed regime not immediately conforming to the norm. In the static regime, friction increases linearly; note that the x-axis of the graph shown is logarithmic, due to the frictional resistance of the two

contacting surfaces until a distinct "break-away"-point occurs, the limiting friction (Pondicherry et al., 2018), and the boundary regime starts. The friction coefficient continues to rise as more and more of the sample is entrained between the surfaces, until enough has been entrained that the surfaces do not touch. The mixed regime seems to be divided into distinct sections: the first part starts at the maximum of the boundary regime and ends with the onset of what the authors of this paper have dubbed the plateau. Indeed, if the obtained friction curves were analysed only in the speed range  $10^{-5}$ - $10^{-3}$  m/s, one could argue that the onset of the plateau is the beginning of the elasto-hydrodynamic regime. The first "hump", or stick-slip event, in the plateau seems to be a characteristic of the measuring system, rather than something related to the actual samples, as it occurs at almost the same speed across samples. Whether this is a property of beer or the measuring system needs to be verified by further research. The plateau ends, and the second mixed regime starts. In the present analysis, this second mixed regime is separated into m1 and m2 based on the logarithmically falling nature of the curve, until it breaks and starts to flatten.

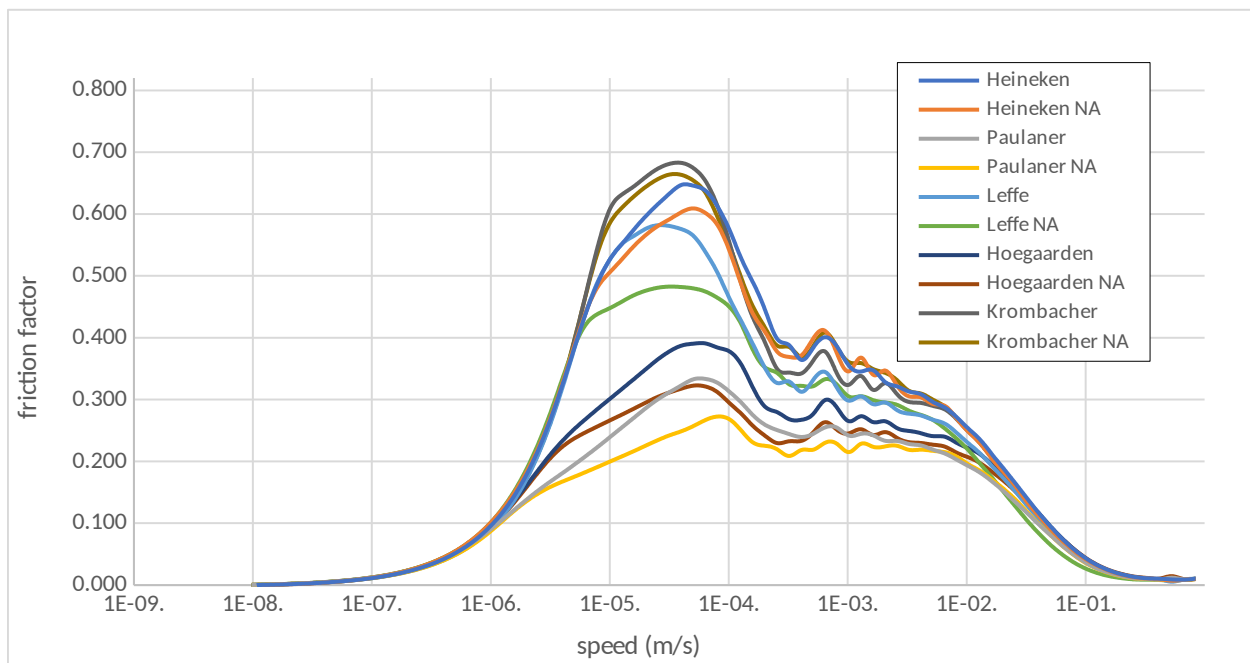


Figure 9: Frictional parameters of the measured beers (error bars indicate standard deviation).

Based on friction curve variables, the beers divide into two clusters, with the wheat beers on one side and lagers/pilsner on the other side (Fig 16). Comparing the two graphs, it is evident that the mixed regimes describe the Hoegaarden, normal and NAB, while Paulaner, normal and to some extent NAB, is described by  $s\_slope$ . It is also evident that viscosity is important, as the beers align themselves on either side along the axis of this variable.

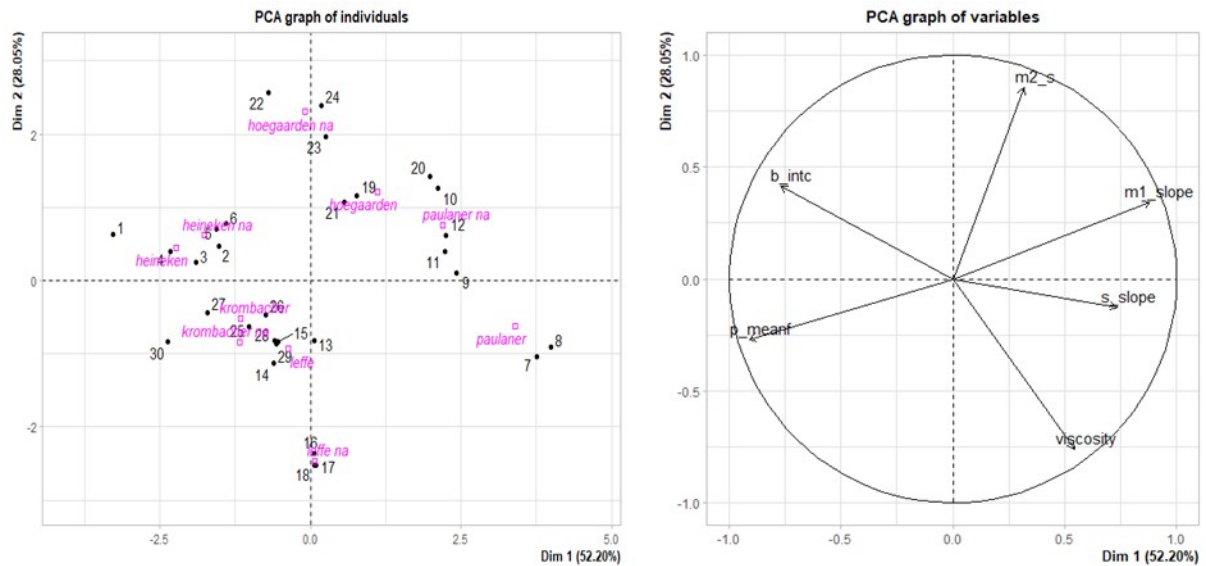


Figure 10: Individuals plot and variables plot of retained tribology variables and viscosity.

### 3.3 Sensory data

#### 3.3.1 Initial analysis

The preliminary question to answer regarding this dataset is its' validity and credibility. Given that the origin of the data is online and the reviews are anonymous, controlling for bias; age, gender, socioeconomic background etc., is impossible, and the purpose and intention; e.g. economic interests, of the reviewer in providing the review is unknown. The maintainers of the website, however, conduct strict quality control to ensure that all reviews are authentic; fraudulent reviews and accounts are deleted, and ratings by brewers or brewer affiliates of their own products are strictly forbidden (Ratebeer.com, 2020).

Any bias introduced as a consequence of effects of number of reviews by reviewers as well as the time of reviewing, e.g. changes in the product over time, were excluded by correlating these two variables with all other relevant variables and calculating Pearson's  $r$  which, in all cases, were either low ( $r < [0.23]$ ) or insignificant ( $p < 0.05$ ) (table 7). Flavour and aroma are highly positively correlated ( $r > 0.8$ ) with overall rating, mouthfeel is highly correlated ( $r = 0.72$ ) while appearance ( $r = 0.51$ ) seem to contribute less to the overall score. This is in line with findings by Van Doorn et al. (2019), who found that appearance has only moderate influence on other sensory rating terms (e.g. flavour, taste, aroma).

Table 7: Correlation matrix (Pearson's  $r$ ) of numeric sensory variables, average number of reviews by reviewers pr beer (rev\_no\_revs) and average year of reviewing pr beer (year). Numbers in bold are statistically significant ( $p < 0.05$ ).

	appearance	mouthfeel	aroma	flavour	overall	rev_no_revs	year
appearance	<b>1.00</b>	<b>0.53</b>	<b>0.50</b>	<b>0.49</b>	<b>0.51</b>	-0.01	-0.04
mouthfeel		<b>1.00</b>	<b>0.63</b>	<b>0.71</b>	<b>0.72</b>	<b>0.11</b>	<b>-0.21</b>
aroma			<b>1.00</b>	<b>0.83</b>	<b>0.82</b>	<b>0.09</b>	<b>-0.18</b>
flavour				<b>1.00</b>	<b>0.88</b>	<b>0.10</b>	<b>-0.21</b>
overall					<b>1.00</b>	<b>0.09</b>	<b>-0.23</b>
rev_no_revs						<b>1.00</b>	<b>-0.34</b>
year							<b>1.00</b>

Given that the average number of reviews by reviewers is  $5377.1 \pm 6427.4$  at its lowest (Leffe, NA) and  $14593.1 \pm 7561.6$  at its highest (Hoegaarden), it is very likely that most reviewers in this study would consider themselves beer-experts or -connoisseurs; however, the level of beer expertise does not necessarily correlate with an increased ability to distinguish between beers, i.e. perceptual abilities, but rather raises the specificity of the sensory terminology (Giacalone et al., 2016; Van Doorn et al., 2020). As part of the aim of this work is to break down an encompassing sensory attribute, mouthfeel, into several specific attributes, the ability to verbalise sensory perceptions is considered as necessary. The selection of reviews by beer-experts does however potentially introduce a certain amount of bias; beer-experts generally preferring stronger, more complex beers and they tend to rate in more extreme terms, i.e. liked beers are rated higher and disliked beers rated lower as compared to a rating by a beer novice (McAuley & Leskovec, 2013). This raises the possibility that the lighter lagers will generally be rated



lower than, e.g. the wheat beers. This is also exemplified by a brief look at the highly rated beers section on ratebeer.com: most are dark beers or sour beers, e.g. lambics, Berliner Weisse, melomels/fruited beers, and there is no mention of pilsners or lagers (Ratebeer.com, 2020). Another source of bias is brand recognition; the possibility that beers from certain producers, i.e. conglomerate breweries, will be rated based on brand recognition and sentiments towards that particular brand rather than sensory quality. In terms of geography, the USA accounts for 23.6% of the reviews followed by Denmark, England and the Netherlands (figure 17).

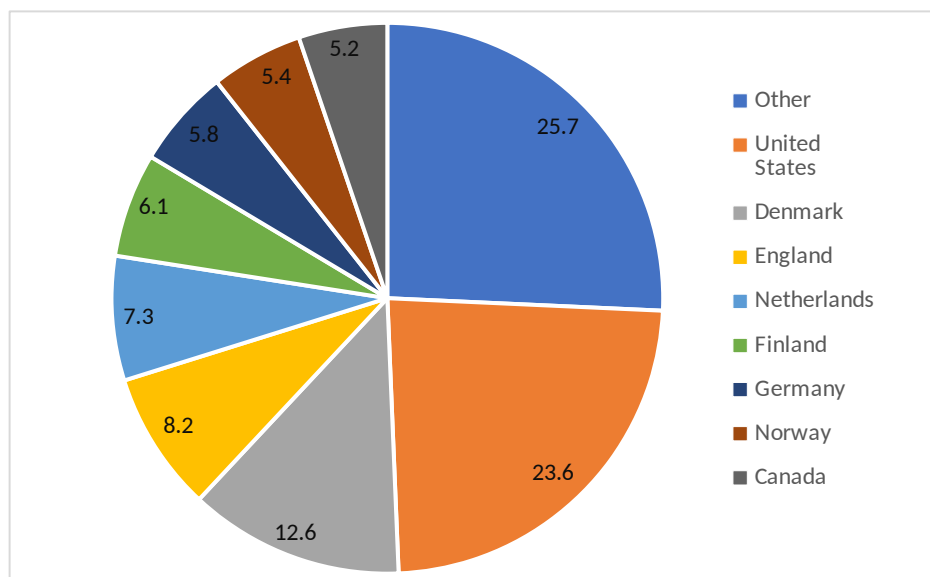


Figure 11: Country of origin of reviewers (%).

All of the non-alcoholic beers are rated similarly and group with Heineken as having a low mouthfeel rating. Paulaner and Hoegaarden both have high mouthfeel ratings with Leffe and Krombacher following. Despite reportedly being brewed with the same ingredients as the alcoholic version, all the NABs have low mouthfeel scores. The three pilsner/lager style beers, in increasing order, Heineken, Krombacher, Leffe, were rated relatively low. Interestingly, Leffe NA with its' low concentration of fermentable sugars (indicating higher degree of attenuation) (Table 8) is the highest rated NAB.

Table 8: Summary of the numeric sensory ratings of the beers as well as the mean number of reviews per reviewer. Mouthfeel and appearance are rated on a scale of 1-5, aroma and flavour on a scale of 1-10 and overall on a scale of 1-20. Different letters in superscript signify significant differences ( $p < 0.05$ ).

Beer	N	Mouthfee				Mean number of	
		I	Aroma	Flavour	Appearance	Overall	reviews
Heineken	191	2.3±0.6 <sup>d</sup>	3.8±1.3 <sup>e</sup>	4.1±1.2 <sup>d</sup>	2.5±0.7 <sup>f</sup>	7.9±2.7 <sup>f</sup>	13925.9±7569.8 <sup>a</sup>
Heineken NA	151	2.1±0.8 <sup>d</sup>	4.1±1.4 <sup>e</sup>	4.1±1.5 <sup>d</sup>	2.7±0.9 <sup>ef</sup>	8.1±2.8 <sup>ef</sup>	8056.2±8548.1 <sup>b</sup>
Hoegaarden	190	3.4±0.7 <sup>ab</sup>	6.8±1.0 <sup>a</sup>	6.7±0.9 <sup>ab</sup>	3.5±0.8 <sup>ab</sup>	13.9±2.0 <sup>a</sup>	14593.1±7561.6 <sup>a</sup>
Hoegaarden NA	73	2.2±0.6 <sup>d</sup>	4.8±1.3 <sup>cd</sup>	4.2±1.4 <sup>d</sup>	2.8±0.6 <sup>ef</sup>	8.2±2.9 <sup>ef</sup>	8729.2±9725.6 <sup>b</sup>
Krombacher	190	2.9±0.6 <sup>c</sup>	5.2±1.0 <sup>c</sup>	5.3±1.1 <sup>c</sup>	3.1±0.7 <sup>d</sup>	10.7±2.2 <sup>c</sup>	12563.4±8079.9 <sup>a</sup>
Krombacher NA	121	2.3±0.8 <sup>d</sup>	4.1±1.4 <sup>e</sup>	4.2±1.6 <sup>d</sup>	2.9±0.9 <sup>de</sup>	8.1±3.4 <sup>ef</sup>	7550.6±9812.3 <sup>b</sup>
Leffe	190	3.2±0.6 <sup>b</sup>	6.4±1.0 <sup>b</sup>	6.4±1.1 <sup>b</sup>	3.4±0.6 <sup>bc</sup>	13.0±2.2 <sup>b</sup>	14566.9±7633.3 <sup>a</sup>
Leffe NA	28	2.4±0.7 <sup>d</sup>	5.1±1.5 <sup>cd</sup>	5.1±1.3 <sup>c</sup>	3.0±0.8 <sup>de</sup>	9.9±3.0 <sup>cd</sup>	5377.1±6427.4 <sup>b</sup>
Paulaner	191	3.5±0.6 <sup>a</sup>	6.8±1.0 <sup>a</sup>	6.9±0.9 <sup>a</sup>	3.6±0.7 <sup>a</sup>	14.0±1.9 <sup>a</sup>	14439±7655 <sup>a</sup>
Paulaner NA	169	2.3±0.7 <sup>d</sup>	4.6±1.2 <sup>d</sup>	4.3±1.2 <sup>d</sup>	3.2±0.7 <sup>cd</sup>	8.8±2.6 <sup>de</sup>	8903.5±9150.4 <sup>b</sup>

### 3.3.2 Text-mining of reviews

The numeric sensory descriptor mouthfeel, although valuable, only holds information on the liking/rating of mouthfeel as understood by each individual reviewer. The guidance from ratebeer.com on the rating of mouthfeel is as follows: "The body of the beer, carbonation and astringency" with no further indications or explanations. In order to further analyse mouthfeel and separate it into more descriptive terms, the individual reviews were analysed by text-mining (section 2.5). An important limitation to the text-mined dataset is that as each word is analysed on its' own, the context in which it is used gets lost. This means that the use of the word watery does not necessarily mean that the beer in question is actually watery, as the connotation of the word can be drastically altered by a conjunction, i.e. not watery (Stavrianou et al., 2007). The basic assumption in this analysis is therefore that the higher the frequency, the higher the likelihood that the word in question is used to directly describe the beer.



side, low mouthfeel rating, with terms such as "watery" and "thin" dominating, while on the right side terms such as "creamy" and "smooth" are more pronounced. Two outliers can be identified; Heineken, although alcoholic, clearly groups together with the non-alcoholic beers, while Leffe NA groups closer to the alcoholic beers than to the non-alcoholic versions. Figure 18 also illustrates the difficulty in directly extrapolating the use of a word with a positive or negative mouthfeel score, as bitterness and bitter seem almost ubiquitous and the terms "watery" and "thin" are relatively frequently used to describe Paulaner and Hoegaarden, even though these beers scored the highest in terms of mouthfeel. Krombacher is an outlier in terms of descriptive mouthfeel terms, being described mainly as "bitter", but also "watery" and "thin", yet scoring higher in mouthfeel than other beers described similarly, i.e. the NABs).

Table 9: Correlation matrix (Spearman's rho) of numeric mouthfeel rating (mouthfeel) and sensory descriptors. Numbers in bold are statistically significant ( $p < 0.05$ ).

	mouthfeel	watery	smooth	bitter	thick	foam	astringen t	sour
mouthfeel	1.00	<b>-0.85</b>	<b>0.71</b>	-0.05	<b>0.65</b>	-0.05	<b>0.71</b>	-0.01
	watery	1.00	<b>-0.71</b>	0.16	<b>-0.68</b>	-0.05	<b>-0.68</b>	0.20
		smooth	1.00	-0.32	<b>0.78</b>	<b>-0.38</b>	<b>0.53</b>	0.26
			bitter	1.00	-0.25	<b>0.68</b>	<b>0.42</b>	<b>-0.64</b>
				thick	1.00	0.05	<b>0.53</b>	-0.14
					foam	1.00	<b>0.39</b>	<b>-0.84</b>
						astringent	1.00	-0.33
							sour	1.00

Table 9 presents the correlation matrix (Spearman's rho) of the descriptive mouthfeel variables. A strong negative correlation occurred between "watery" and overall mouthfeel rating, while "smooth" and "thick" are positively correlated with mouthfeel rating. These results confirm those of Malfliet et al. (2009) who found that non-alcoholic beers with high perceived fullness were generally preferred by a taste panel. As expected, both "smooth" and "thick" are negatively correlated with "watery". The positive correlation between "foam" and "bitter" is also expected because of the well-established relationship between hops bitter compounds and their role in foam stability (Ferreira et al., 2005;

Hughes, 2000). The correlation of "smooth", "thick" and "watery" to "astringency" seems more circumstantial than causal, while the positive correlation (although smaller) of "astringency" to "bitter" and "foam" is expected. The negative correlation of "sour" to "bitter" and "foam" and the non-significant correlation with astringency seems to confirm, rather than disprove, the assumption that "sour" is sometimes wrongly used to describe perceived astringency: if a reviewer describes a beer as sour with the intention of describing astringency, then it is unlikely that the reviewer will also use the word astringent. The negative correlation with "bitter" can be explained by the fact of mutual exclusivity, i.e. if a reviewer describes something as sour with the intention of describing bitterness or astringency, then it is unlikely that all three words will appear in the same review.

### 3.4 Link between sensory and physical and chemical parameters

Comparison of the current results with literature is complicated by a number of factors, namely; 1) tribological measurements and their output are heavily dependent on the system parameters, i.e. the measured sample, the measuring system, and the surfaces used, making comparison between studies challenging at best (Fox et al., 2020; Sarkar & Krop, 2019; Shewan et al., 2019), 2) no published literature involving beers and tribology exist, and 3) the sensory data is uncontrolled and unsupervised, in contrast to many existing studies comparing sensory perception to tribology where trained sensory panels are often used (Sarkar & Krop, 2019).

	s_slope	b_intc	p_meanf	m1_slope	m2_s	viscosity	mouthfeel
mouthfeel	<b>0.59</b>	<b>-0.48</b>	<b>-0.40</b>	0.28	-0.16	<b>0.65</b>	<b>1.00</b>
watery	-0.32	0.24	0.34	-0.26	0.17	<b>-0.49</b>	<b>-0.85</b>
smooth	<b>0.52</b>	<b>-0.46</b>	<b>-0.58</b>	<b>0.45</b>	-0.07	<b>0.58</b>	<b>0.71</b>
bitter	-0.17	0.24	<b>0.75</b>	<b>-0.79</b>	<b>-0.73</b>	-0.05	-0.05
thick	<b>0.52</b>	<b>-0.63</b>	<b>-0.42</b>	0.31	-0.19	<b>0.72</b>	<b>0.65</b>

foam	-0.24	0.15	<b>0.69</b>	<b>-0.68</b>	<b>-0.57</b>	0.01	-0.05
astringent	0.24	-0.30	0.14	-0.30	<b>-0.64</b>	<b>0.61</b>	<b>0.71</b>
sour	0.20	-0.21	<b>-0.60</b>	<b>0.56</b>	<b>0.60</b>	-0.02	-0.01

Table 10: Correlation matrix (Spearman's rho) of tribology variables and viscosity versus mouthfeel descriptors (numeric rating and sensory descriptors). Numbers in bold are statistically significant ( $p < 0.05$ ).

Table 10 presents correlation coefficients (Spearman's rho) between selected friction curve parameters and sensory attributes of the 10 beers. Starting from the top, the numeric mouthfeel parameter is highly correlated with the slope of the static regime as well as viscosity but less correlated with  $b_{intc}$  and  $p_{meanf}$ . It is perhaps not surprising that viscosity is negatively correlated with "watery" and positively correlated with "thick" which is in line with results obtained by He et al. (2016) and Kokini et al. (1977). The static regime is a relatively underexplored area of friction curves, having only recently been introduced (Pondicherry et al., 2018). The slope of the static regime is correlated with the "break-away" point", signifying the beginning of boundary lubrication. The lower the slope, the less energy is needed to commence movement of the tribopairs and thereby the higher the lubricating properties of the beer. Both "smooth" and "thick" are positively correlated with lower friction as can also be evidenced by correlations with  $b_{intc}$  and  $p_{meanf}$ . Using a steel ball and PDMS disk, smoothness has previously been correlated with the mixed and hydrodynamic regime of oil-in-water emulsions (Upadhyay & Chen, 2019). The lower viscosity of the beer might explain a shift towards lower speeds of this sensory descriptor. Of the mouthfeel descriptors, "watery", "smooth", "thick" and "astringent" are correlated with mouthfeel rating, showing that these concepts are heavily weighted by reviewers when rating mouthfeel. A watery or thin mouthfeel has previously been associated with NABs (Krebs et al., 2019) and astringency is generally considered undesirable in beers, being associated with expired or aged beer (François et al., 2006). Astringency is considered a multimodal sensory perception, depending largely on interactions between salivary proteins and plant polyphenols (Laguna & Sarkar, 2017). Astringency has so far been studied using tribology in wine (Laguna et al., 2017) or tea (Rossetti et al., 2009). Laguna et

al. (2017) found no relationship between friction and concentration of wine tannins, which is in contrast to findings by Wang, Olarte Mantilla, Smith, Stokes, & Smyth (2020) who found that tannins and pH were both critical for explaining the sub-qualities "drying" and "pucker" in a model wine system including saliva. In beer, astringency is caused by polyphenols from barley husk or hops (François et al., 2006; Goiris et al., 2014). The mechanisms underlying astringency in beer could be further explored by using tribology and model beer systems, as well as incorporation of saliva. "Foam", "sour" and "bitter" on the other hand, seem not to influence the mouthfeel rating of the beers, but do show some interesting and high correlations with the plateau and mixed regime. Foam depends mainly on barley proteins and hop acids (Evans & Sheehan, 2002). Protein content (albeit the origin is dairy) has previously been shown to be negatively correlated with friction (Sonne et al., 2014), and future endeavours could explore the role of barley proteins and the possible link between foam and friction.

## 4 Conclusion

In conclusion, a reproducible method for measuring the friction profile of beers was developed. Using this tribology method, the beers could be distinguished into those with high and low mouthfeel depending on their alcohol content. A distinct friction profile deviating from the classic Stribeck curve could be observed. It was possible to extract valuable sensory profiles for the selected beers by text-mining online reviews and correlate these sensory profiles with tribological parameters. However, the method of text-mining online reviews for sensory data should be validated by cross-referencing and comparing with a sensory panel.

The two wheat-beers and the Belgian lager were the most favoured beers in terms of mouthfeel, characterised by reviewers as being predominantly "creamy" and "smooth". All the NABs were described as being "thin" and "watery" except for Leffe NA, the highest scoring NAB, which was described mainly as "creamy" and "bitter".

Overall, "smooth", "watery" and "thick" were correlated with parameters at lower speeds corresponding to the static, boundary and beginning of the mixed regime, while at higher speeds, sensory perceptions of "bitter", "foam", "astringent" and "sour" could be discerned.

Several interesting relationships were discerned from the data of the present study, especially the clear (expected) positive correlation of "smooth" and "creamy" with overall mouthfeel rating (and conversely negative correlation with "watery").

Future efforts should look into the correlation between astringency and concentration of barley and hops polyphenols and pH. Additionally, the impact of incorporation of saliva on the friction curves should be investigated.



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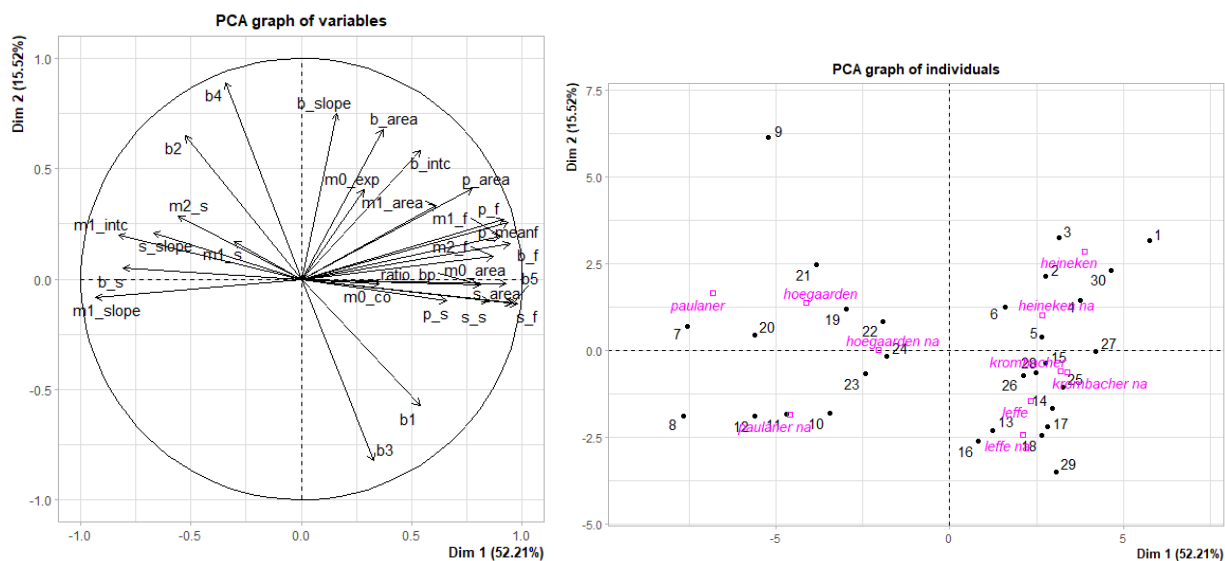
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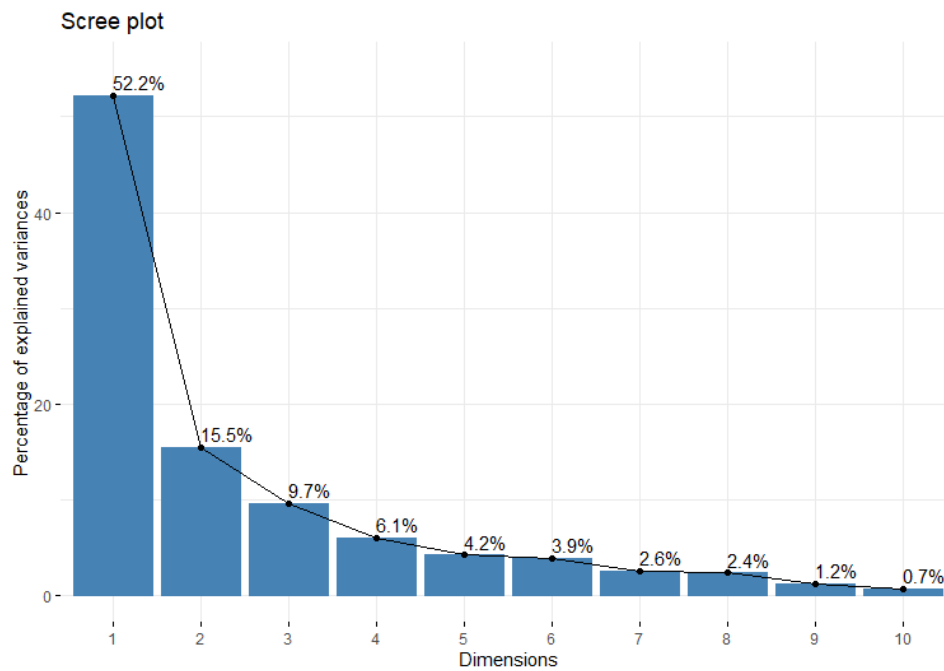
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# 6 Supplementary materials

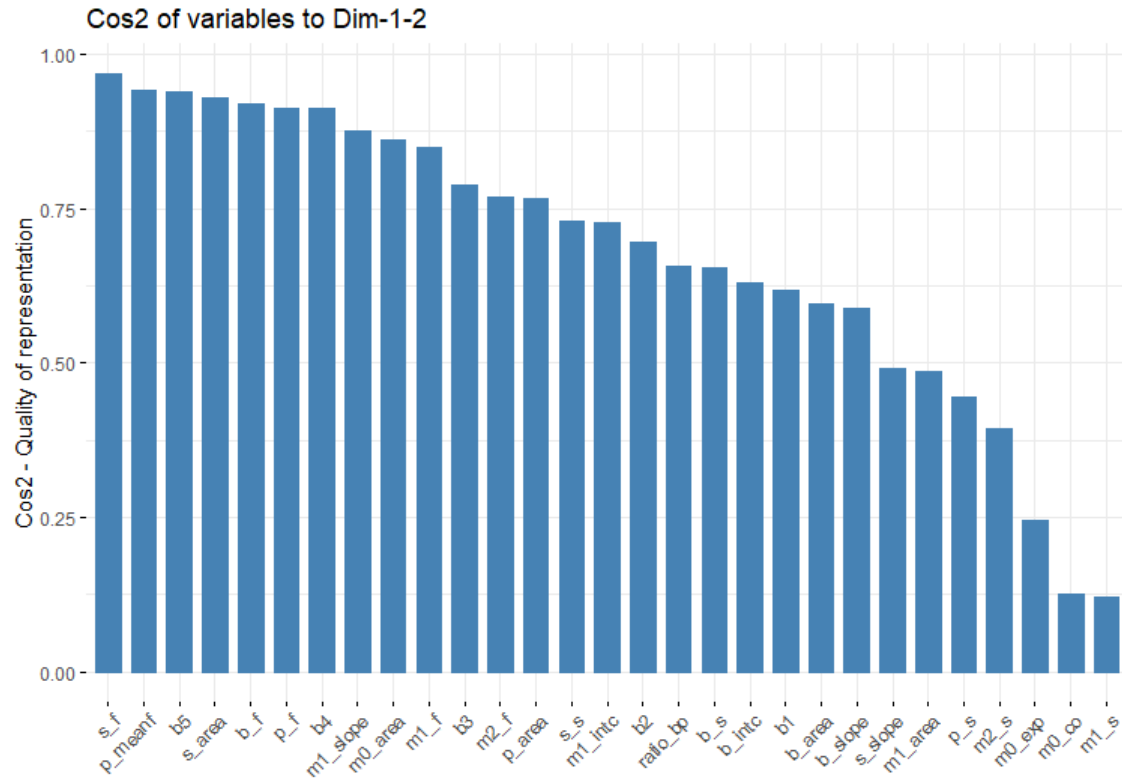


Supplementary figure 1: Variables and individuals of tribology data plotted against PC 1 and 2.

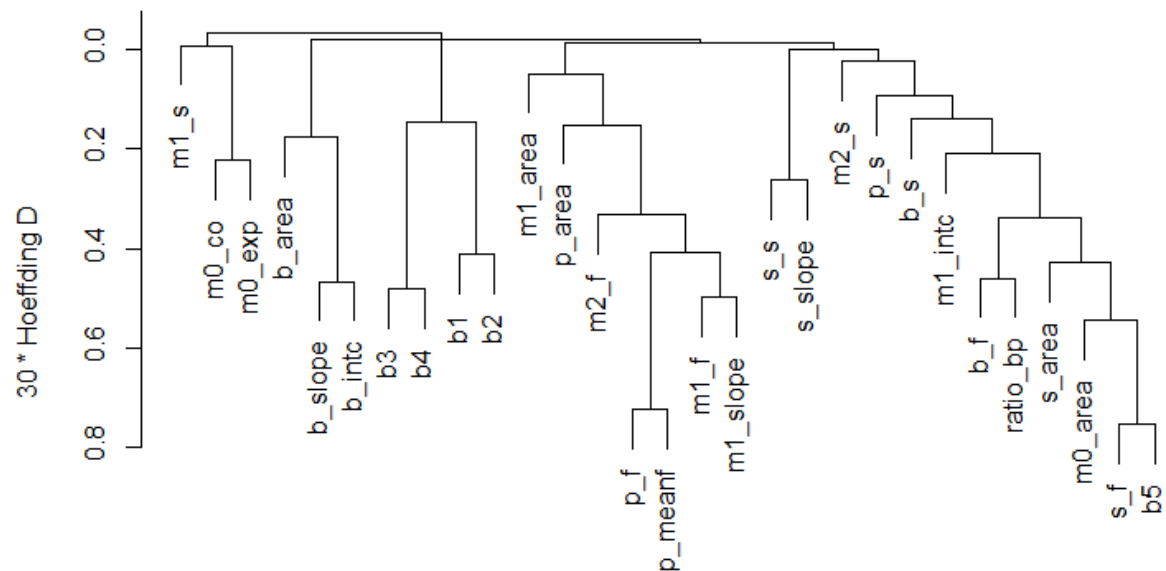


Supplementary figure 2: Scree plot of PCA of all tribology variables.

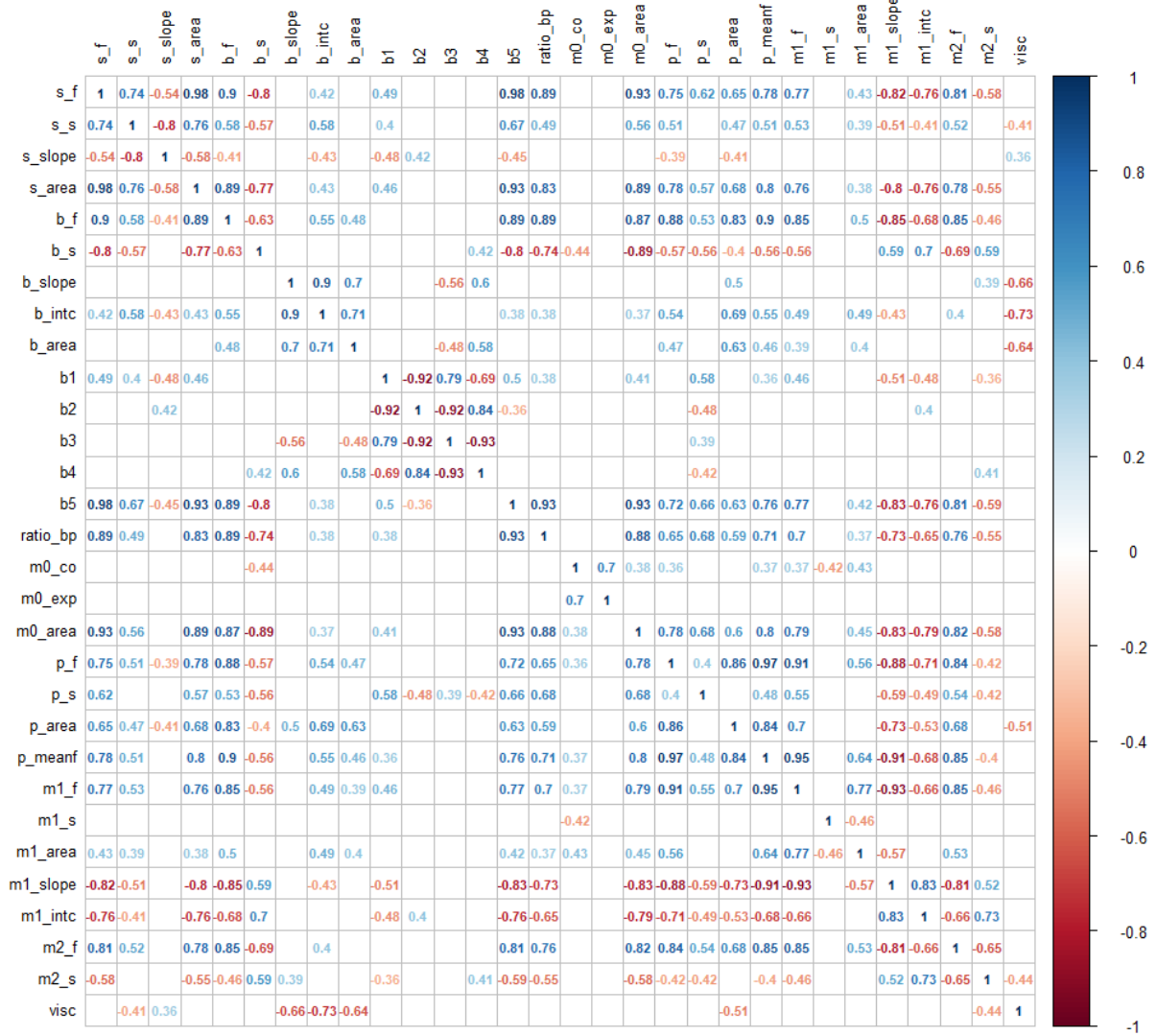




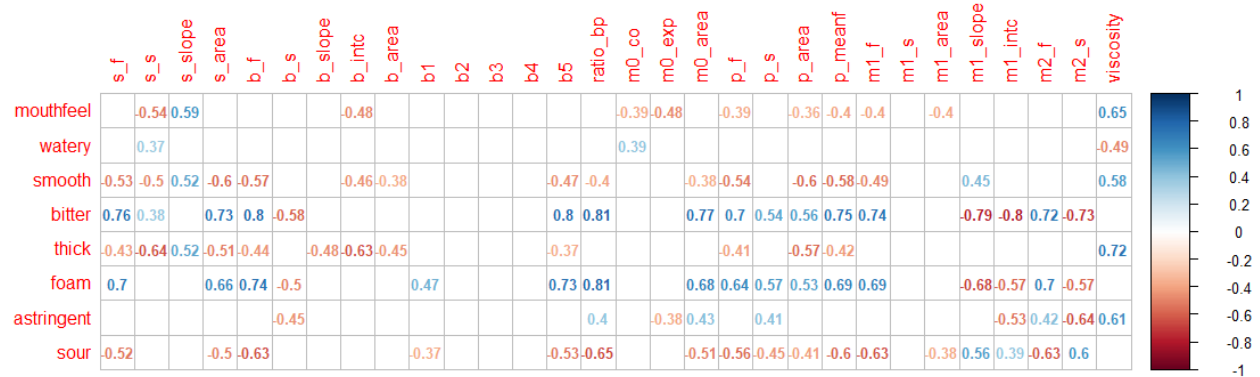
Supplementary figure 3: Quality of representation of tribology variables on PCs 1 and 2.



Supplementary figure 4: Dendrogram of tribology variables using Hoefding's D to test for non-monotonic dependence.



Supplementary figure 5: Correlation matrix (Spearman's rho) of all tribology variables and viscosity.



Supplementary figure 6: Correlation matrix (Spearman's rho) of numeric mouthfeel rating and sensory descriptors vs tribology variables.